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CONSIDERATIONS ON THE RISK OF CHEMICAL CONTAMINATION WHEN IRRIGATTING WITH WASTEWATER

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Abstract: The main reason for heavy metals pollution in agriculture is the use of untreated industrial and municipal wastewater for irrigation, followed by the presence of these inorganic contaminants in products such as fertilizers and pesticides. Heavy metals from irrigation wastewater are easily transferred to the soil–crop–crop consumer system, posing severe threats to environmental sustainability, soil and human health. Heavy metals accumulated in the soil are toxic to microorganisms and disrupt the soil ecosystem. Leafy vegetables tend to accumulate higher concentrations of heavy metals than other parts of the crop. After consumption, heavy metals accumulate in certain human organs and cause serious diseases. Emerging pollutants from the category of pharmaceutical products and pesticides are also found in the waste water, which are very difficult to eliminate through the classic purification procedures, and which are easily transferred and cause disturbances, especially in the aquatic ecosystems. In this paper, aspects related to the route of these contaminants from agriculture and their effects on the environment and health are reviewed.

Keywords: wastewater, heavy metals, emerging contaminants, pharmaceutical products, pesticides

INTRODUCTION

Soil ecosystems are critical to human and environmental sustainability. Many pollutants are currently present in the soil, including heavy metals, organic pesticides, radioactive elements (cesium and strontium compounds), selenium, arsenic, fluorine compounds, and so on. These pollutants enter or remain in the soil for a long time, and once they exceed the soil's self– purification capability, they will directly cause contamination of agricultural soils, potentially resulting in the loss of the production capacity of the soil and to soil being removed from the agricultural circuit.

In any agricultural system, irrigation is essential for obtaining satisfactory and sufficient productions. Irrigation is important for economically successful agriculture in arid and semi–arid regions; also, irrigation is frequently required as an additional measure in semi–wet and wet regions. Wastewater irrigation is simultaneously an effective disposal method and a path of water recovery. Irrigation with wastewater reduces the pressure on fresh water sources and the volumes of effluents discharged into the environment.

Chemical contaminants in wastewater, including heavy metals, pharmaceutical compounds and pesticides, are a cause for concern especially in countries at the beginning of industrial development, where industrial wastewaters enter domestic wastewater streams and natural water streams in larger quantities or even in an uncontrolled manner. When crops irrigated with wastewater are intended for human or animal consumption, the risk translates into the possible introduction of unwanted substances into the food chain; the health effects of heavy metals and pharmaceutical compounds (even more if these categories of contaminants are combined), are not yet fully known (*de Santiago–Martin et al., 2020*).

The World Health Organization has established guideline values for a selection of chemicals that may be harmful in the context of agricultural wastewater use (*WHO, 2006*). In addition, starting with 26 June 2023, the new regulation on minimum requirements for water reuse for agricultural irrigation, established by the
European Commission, is applied in the European Commission, is applied in the European Union (*EC Regulation EU 2020/741*).

HEAVY METALS

Heavy metals are metals and metalloids with densities greater than 5 g/cm3. The most common heavy metals are arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb) and mercury (Hg) (*Herawati et al., 2000*). Cadmium, lead, zinc, and copper are the most prevalent heavy metal contaminants in many countries today (*Yan et al., 2022*). Large amounts of heavy metals are produced and emitted in all industries as a result of rapid urbanization, industrialization and intensification of agriculture. Over the last half–century, the worldwide environment has gained a considerable amount of heavy metals (i.e., over 0.03 million tons of chromium and 0.8 million tons of lead), primarily in the soil (*Yang et al., 2018*). In the European Union, around 3.5 million sites are contaminated with heavy metals, with 0.5 million sites being highly contaminated and in need of remediation (*Mahar et al., 2016*).

The main source of contamination with heavy metals in agriculture is irrigation with untreated industrial or municipal wastewater (or their mixtures), and in some areas even the uncontrolled discharge of these wastewaters that seep into agricultural soils and the groundwater. Untreated wastewater used in irrigation contains high levels of trace elements and heavy metals, being likely to be toxic to plants and obviously posing risks to the environment and human health (*Ungureanu et al., 2020*). In addition, the use of heavy metals in products intended for agriculture (fertilizers, pesticides and other agriculture–based chemicals) is increasingly viewed as a secondary source of pollution in agriculture (*Tchounwou et al., 2012*).

In comparison to potassium and nitrogen fertilizers, phosphate fertilizers contain a significant amount of harmful heavy metals (particularly cadmium and lead) (*Bitew and Alemayehu, 2017*). Heavy metal contamination of soil and crops always occurs in tandem, and it is expected to have consequences on human health. Heavy metals can easily enter the atmosphere, groundwater, soil, and crops, and then the human body by inhalation, ingestion, and skin absorption. Even if these contaminants are present in small amounts (traces), their harmful impacts could disrupt the soil environment. The persistence of heavy metals in the environment is dangerous in the long term, given their long half–life (eg 1460 days for lead and 200 days for cadmium).

Figure1 – Anthropogenic sources of heavy metal pollution of agricultural soils The problem of trace elements and heavy metals in wastewater is mostly attributable to the mixing of municipal, agricultural, livestock, and industrial wastewater streams in the same sewer system in most developing nations. This issue is exacerbated by the flow of untreated industrial wastewaters into natural water bodies, which is permitted under lax pollution control procedures. Mercury, lead, arsenic, copper, cadmium, and manganese are only some examples of potentially dangerous trace metals. It is important to know if traces of heavy metals are: in solution or adsorbed on solids; in organo– metallic or hydroxide forms; in the crystalline structure of suspended materials. Without very accurate distribution data, the development of elimination techniques and guidelines for these harmful elements cannot be effectively designed.

Typically, most treated or partially treated wastewaters have low levels of trace heavy metals, which are within acceptable limits for irrigation water quality. According to *Elgallal et al. (2016*), treated or partially treated wastewater can be used safely for up to a century without adverse effects on soil, crops, groundwater or the food chain. However, several studies have mentioned that after 50–100 years of irrigation with wastewater, the level of toxic heavy metals in the soil exceeded the maximum permissible limits.

Different properties of the soil (pH, texture, type of oxyhydroxides and amount of organic matter), but also the type of plant crops, generally determine the transport of trace metals and essential components (carbonates, phosphates and clay) from the soil to the plants.

Regular use of wastewater for soil irrigation increases heavy metal concentrations in agricultural crops and these metals are subsequently transferred through the food chain to humans and animals, causing potential long– term health risks. Health risks due to heavy metals contamination can be viewed as occupational hazards because chemical pollutants in wastewater can affect the health of farmers who come into direct contact with the contaminated water during crop irrigation. Some heavy metals are carcinogenic, mutagenic, teratogenic, and endocrine disruptors; they have the potential to cause substantial damage even at very low concentrations, while more dangerous ones can cause neurological and behavioral changes, particularly in children (*Mahar et al., 2016*).

Examium (Cd) is not considered an essential element for plants, but is efficiently absorbed by both root and leaf systems. Cadmium has high toxicity and high levels of persistence in

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food and the environment, being the most dangerous heavy metal in the environment (*Perez–Lopez et al., 2008*). Studies conducted in China, Japan and Taiwan by *Carr et al. (2004)* showed that rice grown in soils contaminated by irrigation water containing substantial industrial discharges accumulated high concentrations of cadmium (and other heavy metals). Cadmium exposure can produce a wide variety of acute and chronic effects in humans, such as kidney failure, lung failure, bone damage, and hypertension (*Sun and Li, 2011*). Since one way of human exposure to cadmium is through food consumption, assessing and controlling the amount of contaminated food sources, identifying sources of contaminants as well as modifying or eliminating them play a significant role in human health and longevity.

Chrome (Cr) is considered as an environmental pollutant released into the atmosphere mainly due to its intensive use in heavy industries. Chromium is a well–known human carcinogen, and numerous respected institutions worldwide have proven lung cancer as a result of the exposure to this contaminant (*Tirger et al., 2008*).

Copper (Cu) plays an important role in photosynthesis, respiration, carbohydrate distribution, nitrogen reduction and fixation, protein metabolism and cell wall metabolism. Copper regulates the creation of DNA and RNA, and a lack of it severely limits plant reproduction. It is difficult to define and specify the amount of copper in the soil that has harmful effects on plants. In general, before phytotoxic symptoms appear, the degree of copper buildup in plants poses a risk to human health.

Iron (Fe) uptake generally depends on soil pH, calcium and phosphorus concentrations, and ratios of several heavy metals. High soil iron concentrations can cause phytotoxic effects in acid, low phosphorus, acid sulfate, and flooded soils.

Lead (Pb). Airborne lead is readily taken up by plants through leaves, from where it is absorbed into plant tissues. Soil lead is not easily translocated into the edible parts of plants. Soil contamination from agricultural operations has been shown to have a minimal effect on plant lead concentrations. Vegetables cultivated in high lead concentration areas, such as urban and industrial areas, may represent a health risk to

those who consume them, because lead absorbed by the human body can harm organs like the heart, bones, and nervous system (*Bruce et al., 2012*).

The different chemical and biochemical transformations that heavy metals can undergo in the aquatic environment deserve special attention. Chemical transformations can affect the bioavailability or toxicity of heavy metals, which can be either enhanced or reduced. Knowledge of the mechanisms underlying the physical, chemical, or microbial transformations of heavy metals is often essential to understanding the health effects of these contaminants.

Farrag et al. (2016) studied the translocation of some heavy metals from soils irrigated with wastewater in some crops such as cabbage, onion, garlic and wheat. Their results showed that Cd, Cr, Cu, Ni, Pb and Zn accumulated in the edible parts of the crops and the concentrations of these heavy metals varied greatly depending on the type of agricultural crop.

The accumulation of even small amounts of metals in soil, plants and food products is a challenge in long–term wastewater irrigation areas due to chronic exposure of consumers to food products from crops irrigated with wastewater containing of heavy metals. Several studies have shown that plants irrigated with wastewater can absorb and accumulate potentially toxic elements in concentrations higher than the maximum permissible limits, with serious implications for public health. Long–term consumption of vegetables potentially contaminated with toxic elements can lead to the continuous accumulation of toxic metals in the kidneys and liver in humans, causing disruptions in the physico–biochemical processes (*Mahmood and Malik, 2014*).

Wastewater used for irrigation has been identified as the potential source of heavy metals such as Cd, Cu, Ni, Cr, Pb, and Zn in the soil, plants, and food products (Table 1). Certain food crops pose greater risks of transferring the heavy metals with which they are contaminated, into the human body. In the case of irrigation with industrial effluents, the risk of contamination with heavy metals and trace elements increases.

Information about different uptake levels of heavy metals by different agricultural crop types is useful to determine which vegetables should be grown in areas known to be contaminated with specific pollutants. However, when recommending a particular crop, several context–specific factors must be considered, and not all factors are based on consumer health, but also on market value and growing conditions.

Table 1. Concentration of heavy metals in wastewater, soil and vegetables, in relation to transfer and bioaccumulation factors (Khalid et al., 2018)

Since trace elements and heavy metals in wastewater can be toxic to plants at levels below those that pose a significant risk to human health, a certain level of natural protection could also result, because no crop will thrive when irrigated with highly toxic water, and farmers will abandon crops that do not grow satisfactorily before exposing themselves to significant health risks through consumption of those crops (*Kilelu C.W., 2004*).

The uptake of heavy metals varies by plant species and different plant parts (growing above or below ground). In other words, chemical pollutants can be retained on the surface of crops and vegetables that grow above ground, in addition to being absorbed through the roots. In general, the risk of contamination with potentially toxic elements is higher for vegetables that have consumable plant parts below ground than those above ground (*Khalid et al., 2018*). Concentrations of heavy metals accumulated in roots are usually higher than in leaves. Leafy vegetables tend to accumulate more easily the heavy metals, but monocots such as rice accumulate a higher concentration of heavy metals in their roots *(Nabulo G., 2002)*. Increased metal deposition in roots and shoots can be useful or poisonous depending on the type of edible component of the vegetable. For leafy plants, heavy metal deposition in roots is

beneficial, whereas for tuber vegetables, significant translocation to shoots is desired. Heavy metal concentrations in plant tissues increase with metal concentrations in irrigation water, and this problem can only be solved by proper wastewater treatment.

Pandey et al. (2012) found that while root vegetables showed higher uptake of heavy metals compared to leafy vegetables and fruits irrigated with wastewater, when atmospheric deposition inputs were considered along with wastewater irrigation, leafy vegetables showed higher levels of contamination.

Several studies have reported high accumulation (above the toxic limit) of potential toxic elements in various edible parts of crops/vegetables worldwide. A study by *Chopra and Pathak (2015)* reported the accumulation of Pb, Cu, Zn, Ni, Cd and Cr in *Beta vulgaris*, *Phaseolus vulgaris*, *Spinacea oleracea* and *Brassica oleracea*. *Mason et al. (2011)* observed the accumulation of Zn, Cu, Mn, Cd, Pb, Ni, Fe and Cr in *Zea mays* (maize) culture irrigated with wastewater and fertilized with sewage sludge. *Jamali et al. (2007)* reported the accumulation of Cd, Cu, Cr, Ni, Pb and Zn in vegetables irrigated with mixtures of wastewater and sewage sludge*. Kiziloglu et al*. *(2008)* found increases in Cu, Fe, Mn, Zn, Pb, Cd and Ni concentrations in red cabbage and cauliflower crops irrigated with wastewater.

În mod similar, *Balkhair and Ashraf (2016)* found that the level of Cr, Pb, Ni and Cd in the edible parts of okra was higher than the safe limit with levels of 63%, 28%, 90% and 83% respectively in the samples, leading to the conclusion that irrigating the okra crop with wastewater containing potentially toxic elements is not safe for human health. *Khan et al. (2008)* found that leafy vegetables have a higher transfer of Cd, Cu and Ni from soil to plants. *Singh et al. (2010)* compared a range of vegetables contaminated with heavy metals and found the highest concentrations in cabbage, eggplant and leafy vegetables such as lettuce and spinach, compared to other vegetables such as some varieties of squash. A study conducted by *Dickin et al. (2016*) found that compared to other crops, the accumulation of heavy metals even at low concentrations in rice and wheat crops poses a greater risk to humans because these crops are consumed in larger quantities in many diets.

Currently, worldwide, different methods of remediation of soils contaminated with heavy

metals have reached the stage of technological maturity and are successfully implemented, while other methods are in the research–development stage.

Figure2 – Anthropogenic sources of heavy metal pollution of agricultural soils (Ashraf et al., 2019)

CONTAMINANTS OF EMERGING CONCERN

Contaminants are considered "emerging" when they have a new source, a different route to people, or novel treatment options (*Gogoi et al., 2018*). Emerging contaminants (ECs) are chemicals, either synthetic or naturally occurring, or microorganisms that are not widely monitored in the environment but have the potential to enter it and have known or suspected detrimental ecological or human health impacts. Emerging contaminants include many micropollutants found in municipal, industrial and agricultural wastewater, such as pharmaceutical products (antibiotics, steroid hormones, x–ray media etc), personal care products, pesticides, insecticides, surfactants, detergents, dyes, polymers, plastics, phthalates, flame retardants, industrial additives.

Figure3 – Sources of emerging pollutants and their routes in the environment (Vasilachi et al., 2021)

Exposure to emerging contaminants has a negative impact mainly on the aquatic environment and animals in direct contact with

polluted water (mainly surface water) (*Ungureanu et al., 2019*). Released into the environment, emerging contaminants have ecotoxicological effects on aquatic and terrestrial organisms (feminization of aquatic organisms, bacterial resistance, endocrine disruption). In addition, all these emerging chemicals reaching have many negative effects on human health.

Figure4 – Negative consequences on human health due to emerging contaminants (Kumar et al., 2022)

Although the risks associated with emerging contaminants in treated wastewater used for irrigation are still controversial, some studies have argued that these contaminants are unlikely to pose a serious threat to groundwater, soil, or human health as a result of their application in agriculture. However, there is still a significant lack of studies on the prevalence and fate of emerging contaminants as a result of agricultural wastewater reuse, in terms of their potentially negative effects on the terrestrial ecosystem, uptake by crops and potential impact on human health. through the food chain (*Elgallal et al., 2016*).

EXECUTE: Pharmaceutical compounds for human and **veterinary use**

Pharmaceutical compounds (antibiotics, steroids, contraceptives, analgesics, endocrine– disrupting chemicals, anti–cancer agents, beta– blockers, lipid regulator agents, anti– inflammatory drugs, anticonvulsants, contrast agents etc) are being used in human and veterinary medicine on a daily basis.

Veterinary antibiotics are widely employed in the prevention and treatment of animal infections, but they are also widely utilized to increase productivity, typically in feed and water, which greatly outweighs their usage as animal therapies at the moment. Antibiotics cannot be fully metabolized, so their residues are found in household wastewater, medical wastewater, livestock and poultry farming wastewater, and, in some cases, in antibiotic production wastewater. The use of veterinary antibiotics as animal growth boosters is prohibited in the European Union due to the numerous environmental concerns (*Ungureanu et al., 2019*).

Steroids are estrogens, androgens, glucocorticoids and progestagens. Natural and synthetic steroids are used in human daily life for contraception and therapy, as well as in livestock production to prevent and treat diseases, boost growth and productivity, and manage animal reproduction. The European Union restricted the administration of steroid hormones for farmed animal fattening due to the negative human health impacts of hormone residues in animal foods (*Ungureanu et al., 2019*). After being assimilated in the human or animal body, these substances are eliminated through metabolic processes in the environment.

In recent years, different concentrations of pharmaceutical compounds, which have negative effects on the environment (especially on the aquatic environment, but also on soil and sediments) have been detected in numerous wastewater flows and natural water bodies. Once in the environment, pharmaceutical products can undergo natural processes such as biodegradation, sorption or dilution, by which their concentrations in water or soil are reduced, or they can be taken up by aquatic organisms and plants. In the case of pharmaceuticals, the transformation products resulting from these processes are most often more soluble and polar than the parent compound and therefore more mobile. The European Medicines Agency specifies that any traces of transformation products exceeding 10% of the concentration of the original compound must be investigated to determine the possible effects on ecosystems.

It was found that there is a slow accumulation of veterinary pharmaceuticals transferred from treated wastewater into plant tissues. The concentration of veterinary pharmaceuticals contained in plants depends on the type of chemical agent, when the wastewater is applied as irrigation water, the type of wastewater and the season (*Christou et al., 2017*). Greater adsorption of chemicals in soil was observed during winter. Although veterinary pharmaceuticals carried into soils irrigated with treated wastewater do not pose a high risk to terrestrial organisms, the concentration of these

products must be constantly monitored to trace their movement into the soil layers (*Biel–Maeso et al., 2018*).

It is highly likely that emerging pollutants pose many threats to soil and human health, but long– term studies are still needed to test this hypothesis. Pharmaceutical chemicals in untreated wastewater used for irrigation might pose a threat to agricultural land because their buildup in the soil can impact microorganisms and soil worms for many years, promoting the development of antibiotic–resistant microbes.

Pharmaceuticals can then be carried from the soil to plants and reach the food chain, potentially endangering the human health. There is still a chance that these chemical compounds will be discharged from the soil into groundwater or surface water, creating a risk to aquatic life biocenosis. Detection of low concentrations of pharmaceuticals is challenging and expensive, requiring the use of specialized analytical techniques such as solvent extraction and ultrasound in conjunction with liquid chromatography (*Montemurro et al., 2019*).

The most effective processes in the removal of pharmaceutical compounds from wastewater proved to be the advanced oxidation processes (particularly Fenton oxidation). However, even if advanced wastewater treatment processes are used, often traces of pharmaceutical products are still found in the effluents, which will end up being eliminated in the environment (transferred to surface water bodies, they pose significant risks to aquatic organisms and humans).

▓ **Pesticides**

Pesticides, a well known group of toxic organic chemicals with widespread use in agriculture and livestock production, have the primary function to boost food output while decreasing weeds and pests. Considering that pesticides have been used for decades and often applied in excess, they are increasingly being detected in wastewater and natural water bodies.

It has been reported that around 95% of pesticides used up to this point did not reach the target insect but were instead deposited in their surrounding habitats (*Nawaz et al., 2021*).

Conventional wastewater treatment processes are not effective in eliminating pesticides, so they are often found in wastewater treatment plant effluents (*Farias et al., 2023*). As partially treated and untreated wastewaters are frequently discharged into agricultural canals, ACTA TECHNICA CORVINIENSIS – Bulletin of Engineering | e–ISSN: 2067 – 3809 Tome XVII [2024] | Fascicule 2 [April – June]

these chemicals must be considered when developing wastewater reuse guidelines.

To some extent, pesticides can be destroyed in the natural environment by microorganisms' metabolic activity or by chemical degradation, but many recalcitrant pesticides and their metabolites are stable over time in the environment. The potential impacts of pesticides are a cause of obvious concern for farmers and consumers, since the reuse in irrigation of waters containing pesticide residues can negatively affect sensitive species, and human health. Pesticides in aqueous systems are extremely dangerous to aquatic organisms and the ecosystem since they are classed as possible endocrine disrupting agents (*Goh et al., 2023*). In terms of human health, long–term exposure to the active components of certain pesticides and their metabolites has been linked to chronic consequences such as birth abnormalities, organ damage, cancer, nervous system diseases and neurotoxicity (*Meftaul et al., 2020*). Moreover, it is well established that pesticides have the potential to contaminate groundwater due to runoff and leaching processes. Pesticides in drinking water sources are viewed as a possible source of health issues, even at low levels of contamination, due to their functions as endocrine disruptors (*Suwannarin et al., 2021*).

CONCLUSIONS

Heavy metals that contaminate agricultural soils are toxic; they have long biological half–life and are difficult to biodegrade. Cadmium, lead, zinc, and copper are the most prevalent heavy metal contaminants in many countries today.

Heavy metal pollution has serious negative effects for human health and the environment. Some heavy metals are toxic even in trace concentrations, and can cause a variety of diseases, the most affected segment of the population being children. Agricultural workers are frequently exposed to contamination with heavy metals, because they come into direct contact with contaminated soil, with wastewater used for irrigation and with harvested vegetables.

The reuse of wastewater as a substitute for freshwater in agriculture can be helpful to the environment and agriculture, but only if correct treatment, planning and management operations are employed,so as to minimize the risks to the environment and human health. **Acknowledgement**

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