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# Wastewater Reuse for Irrigation — Practices, Safe Reuse and Perspectives

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#### 1. Introduction

#### 1.1. Wastewater for irrigation and its role in sanitation and human health

Human impact on water bodies has become relevant since water withdrawal, especially for agriculture, has resulted in overexploitation of rivers, lakes and aquifers. In most countries agriculture represents by far the largest use of water and worldwide, it represents about 70% of total withdrawal and 90% of water consumption [1]. To address this problem wastewater reuse has proven to be an alternative to reduce anthropogenic impacts [2]. In addition, raw wastewater reuse in agriculture is a valuable tool available to developing countries to control pollution and tackle the challenge of increasing food production in water scarce areas. The benefits of reusing water in agriculture are many and beyond doubt: it saves considerable amounts of first-use water that may be assigned to critical uses; it provides nutrients that may substitute chemical fertilizers, increasing soil fertility and crop yield, and reducing production costs; it makes it possible to expand agricultural land in arid areas; it is a relatively cheap disposal method for raw wastewater; and it may avoid pollution of surface water. In addition, it has been demonstrated that the health risks of reusing water in agriculture are minimal as long as its biological quality meets established criteria [3, 4].

However, it should be considered that there are potential negative impacts that may arise, such as soil salinization as well as soil and groundwater pollution with metals and organic compounds. In addition, the use of raw wastewater for crop production poses health risks due to its microbial content, especially bacteria, viruses and parasites, which produce a wide range of diseases since many of them may survive on the environment for long periods of time.



The main issue associated to wastewater reuse is related to public health and infection risks, either real (produces a disease) or potential (transmits the infection but the disease does not develop). Infection rates may be high, low or minimal, depending on the type of pathogen, the infective dose, and the susceptibility of the affected person (host). According to epidemiological studies over the last 20 years, when untreated wastewater is applied to land for crop production, there exist real infection risks caused by pathogens. To reduce such risks, control actions must be implemented, such as treating wastewater to comply with regulation limits; developing fast, cheap, easy, and efficient detection techniques; breaking the disease-infection cycle with medical treatment; and developing education campaigns for the population [4-6].

### 2. International and regional guidelines and country regulations for treated wastewater reuse in irrigation

Globally, wastewater reuse has become significant and this has encouraged many countries to develop local regulations to control water quality for reuse with the aim of reducing health and environmental risks. Due to different geographic, economic, and social characteristics, development of such regulations has been gradual and dissimilar among countries. Developed countries have worked on these regulations for several years and among them, the United States of America, and in particular the state of California, applied the first regulations on agricultural reuse in 1918 [7]. As a result, California has one of the most strict and complete regulatory frameworks. In addition, the United States Environmental Protection Agency (US EPA) has developed regulations and criteria for water reuse, which are used as a reference by many countries. Moreover, the creation of international organizations has led to the publication of general recommendations. The World Health Organization (WHO) published a series of four volumes that include information about agricultural irrigation and reuse in aquaculture [8]. At the same time, the Food and Agriculture Organization of the United Nations (FAO) has its own guidelines that are similar to those of the WHO [9]. WHO proposes limits for indicator bacteria (total or faecal coliforms) and helminth eggs when wastewater is reused for agriculture, considering that helminth eggs are highly resistant to treatment process and common in the environment.

In Europe, Mediterranean countries have detailed legislation about this topic, while countries with high water availability, such as Germany or the United Kingdom, do not regulate reuse as it is seldom practiced. Spain sets limits on faecal coliforms and nematodes based on their 1985 Water Law.

In the Americas, countries like Brazil, Costa Rica, Chile, and Mexico have made progress in developing regulations focused mainly on restricting water reuse based on microbial content, including indicator bacteria, helminth eggs and some metals, allowing organic matter and nutrients that are beneficial for agriculture to be used on land.

In Africa, despite the fact that several countries face a water crisis, many of them have lax or non-existent regulations and thus wastewater reuse is practiced uncontrolled. However, countries like Tunisia have detailed guidelines that include physicochemical and biological parameters, as well as heavy metals, an approach shared by some Mediterranean countries like Saudi Arabia, Israel, and Jordan [9].

In contrast, countries like Palestine, Libya, and Afghanistan, which have low water availability, have not been able to develop their own standards due to political conflicts and generally use either FAO or WHO guidelines. Finally, Indian regulation is outdated (1974) and limits just a few parameters, even though it has the world's second largest population.

Usually, most regulations for reuse in irrigation establish limits on one or more of the following parameters: indicator bacteria (total or faecal coliforms), helminth eggs (intestinal worms), nematodes (a subgroup of helminthes), organic matter (as biochemical oxygen demand, BOD), dissolved and suspended solids, and heavy metals.

#### 3. Successful cases of wastewater reuse for irrigation

Demographic growth and economic development of emerging American countries has promoted the implementation of several agricultural reuse projects, some of which are summarized below.

In Mendoza, Argentina, an area known as Campo Espejo used to be irrigated with raw wastewater (2,000 ha) but currently, 129,600 m<sup>3</sup>/d originating from stabilization ponds are supplied for the irrigation of 1900 ha [10].

Chile has several successful reuse projects such as the Maipo and Maipocho regions where 130,000 ha are irrigated [11]; Antofagasta where about 20,000 m<sup>3</sup>/d of treated water are produced and 65 ha are irrigated; and Santiago de Chile with 110,000 ha that use reclaimed water mixed with first-use water [10].

In Mexico, the central and northern part of the country, where 80% of the population live, is considered arid or semi-arid, and cities like Ciudad Juarez irrigate 26,000 ha with approximately 400,000 m<sup>3</sup>/d of reclaimed water. Additionally, the Mezquital Valley, covering more than 90,000 ha, is one of the largest areas in the world where agricultural reuse is practiced [12] and where a large wastewater treatment plant (35 m<sup>3</sup>/s) is under construction to improve water quality for irrigation.

In South America, Peru encompasses a number of different biomes that range from Amazon rainforest (more than 50% of the country) to the west coast, an area with low precipitation where most of the population lives, and where small irrigation projects like San Agustin (535 ha) and Tacna (738 ha), have been developed [10].

#### 4. Case study: Tula Valley, Mexico

The Tula Valley (also known as Mezquital Valley) is one of the largest areas irrigated with untreated wastewater in the world. The fact that it has been receiving wastewater since the late XIX century, and specifically for irrigation since the early XX century, makes this a unique site with regard to wastewater reuse. A large number of studies have described several processes and phenomena that emerge out of this practice, including incidental aquifer recharge, increase in crop yield, and treatment of the wastewater by the soil (in the same way as a soil-aquifer treatment system, SAT).

Concurrently, other studies have found an increase in health risks after using untreated wastewater in agriculture due to the presence of pathogens, heavy metals, and organic compounds in soil and even in groundwater. This section summarizes the history and current situation of the Tula Valley to share some lessons learned for more than 100 years of water reuse.

#### 4.1. Metropolitan Area of the Valley of Mexico and Tula Valley

The Metropolitan Area of the Valley of Mexico (MAVM) has a population of 21.2 million and covers the Federal District (Mexico City) and 60 surrounding Municipalities (59 located in the State of Mexico and 1 corresponding to the State of Hidalgo). Water use is estimated to be 82 m³/s which includes 91% first use water and 9% reclaimed water. The rate of wastewater generation (including collected precipitation) ranges from 52 to 300 m³/s depending on the season, and it is conveyed to the Tula Valley. The reason for this practice is that Mexico City is located in an endorheic (closed) basin from which wastewater and excess precipitation needs to be transported to avoid flooding. On average, 60 m³/s are sent by gravity or pumping to the Tula Valley via four artificial exits (Figure 1): the Tajo de Nochistongo (deep cutting through Nochistongo Hill; 1607-1789); the Tequixquiac Tunnel (1900); the New Tequixquiac Tunnel (1955) and the Central Emitter (1975). Currently, a fifth exit (East Emitter) with a capacity of 150 m³/s is being constructed along 62 km and will be operating by 2016.

The Tula Valley is located 100 km north of Mexico City, with an elevation that ranges from 1,700 mamsl in the north, to 2,100 mamsl in the south. The climate is semiarid with an annual precipitation of 550 mm (national average is 790 mm) mainly between May and October [10] which contrasts with evapotranspiration that reaches 1,524 mm/y.

Originally, soils were low in organic matter and nutrients, and with such low precipitation, productivity was also low. By the end of the XIX Century, the Tula Valley started receiving raw wastewater from the MAVM. The first documented use of wastewater for irrigation in areas closed to the Salado River was reported around 1896. However, officially, reuse initiated in 1889 when wastewater was used to produce energy at the Juandhó and La Cañada hydroelectric plants [13], and in 1912 for irrigation [14].

As wastewater generation gradually increased, the irrigated area grew from 10,000 ha in 1920, to 80,888 ha today (Figure 2). This means that about 1,350 Mm<sup>3</sup>/y are transported a distance of 98 km to the Tula Valley. Once in the Valley, wastewater is distributed through a complex hydraulic system that includes six storage dams (combined capacity: 347 Mm<sup>3</sup>), 323 km of main distribution canals, 264 km of lateral distribution canals, and 101 km of agricultural drains. Year-round availability of wastewater, as well as its organic matter and nutrient content which

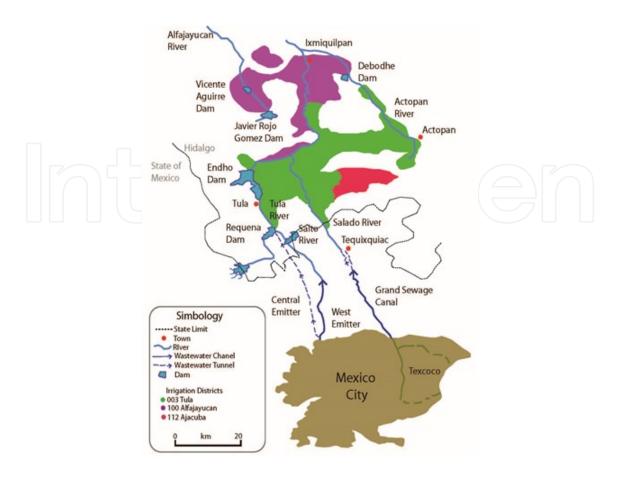


Figure 1. Wastewater transport and use in the Tula Valley.

act as fertilizers, allowed the development of three important Irrigation Districts (ID 003, 100, and 112) that today cover more than 85 thousand hectares [15].

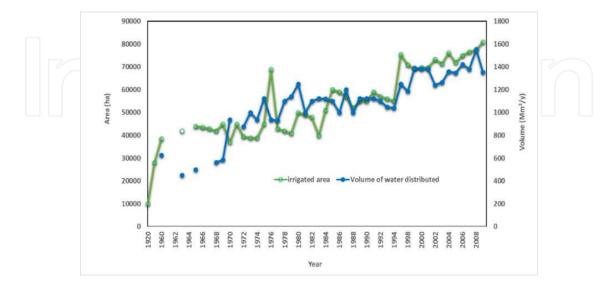


Figure 2. Growth of area irrigated and volume of water distributed since 1920 (adapted from [16], with data from [15]).

#### 4.2. Water reuse in the Tula Valley

The gradual growth of the productive area in the Tula Valley allowed the differentiation of regions with various irrigation ages (Figure 3). Irrigation was first performed in the south-central area of the Valley [14]. Subsequently, when the City expanded and had to dispose of a larger amount of combined wastewater, the irrigation area grew towards the north and the east following the Tula River. Subsequent expansions of the Irrigation Districts carried wastewater to the east and southeast (ID 100), where reuse had started by 1970 [16].

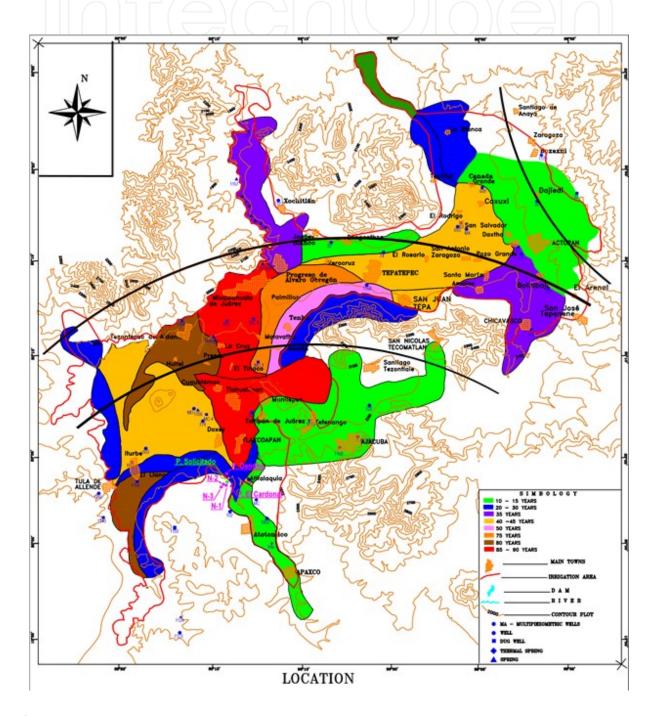


Figure 3. Areas under irrigation for different lengths of time (adapted from [16]).

Recently, wastewater was taken to the far east of the Valley, close to the Ajacuba Municipality (ID 112). As a result, the quality of irrigation water exhibits spatial and temporal variations. For example, in the southern part of the Valley, approximately 10,000 ha are irrigated with raw wastewater, while about 35,000 ha in the central and eastern regions receive diluted wastewater (80% wastewater and 20% river water/precipitation from the Taxhimay and Requena dams) with a different composition.

On the other hand, 25,000 ha located in the far west region of the Valley are irrigated with partially treated wastewater after being stored in the Endho dam which acts as a large settling tank with a hydraulic retention time of up to three months. Finally, the northern areas use well water or return flows and are mainly utilized for growing vegetables and fruits [17].

#### 4.3. Water quality

#### 4.3.1. Microbiological indicators

The potential risk of bacterial, viral and parasitic diseases that can be transmitted through the human-water-soil-crop-human cycle constitutes the greatest problem associated with the use of wastewater in agriculture, for human consumption or for other uses. Thus the study of their removal is especially relevant in developing countries where they represent the higher risk of disease.

The presence of microorganisms can be a serious cause for concern. For example bacterio-phages (viruses) are resistant in the environment, have been shown to have the capacity to penetrate and reach confined aquifers, and have low infective doses [7]. Cysts of *Giardia lamblia* can survive and remain active for months or even years, are resistant to chlorination, have a low infective dose and can have serious consequences for vulnerable individuals [18]. Helminth eggs also survive for long periods of time in the environment, have a low infective dose and their incidence in Mexico is very high [19]. Faecal coliforms are a universally accepted indicator of faecal contamination and have been found to migrate through soil [20].

Wastewater from the MAVM reaches the distribution system at the Tula Valley without any treatment and thus it contains a high concentration of faecal coliforms (between 10<sup>5</sup> to 10<sup>8</sup> colony forming units, CFU/100 mL), *Streptococcus faecalis* (10<sup>2</sup>-10<sup>6</sup> CFU/100 mL), *Clostridium perfringens* (10<sup>3</sup> to 10<sup>6</sup> CFU/100 mL), somatic bacteriophages (10<sup>2</sup> to 10<sup>6</sup> plaque forming units, PFU/mL), *Giardia* spp. (450 to 10,000 cysts/L), and helminth eggs (1.8 to 23 helminth eggs/L) (Table 1). This quality has shown little temporal variation but the concentrations registered pose a significant health risk in areas under irrigation, according to WHO [4].

To evaluate temporal and spatial variability along the Valley, samples from different regions have been taken for microbial analyses during the wet and dry seasons. Zone 1 (south) uses raw wastewater for irrigation and has shown higher concentrations of biological indicators than Zones 2 or 3 which correspond to areas with some sedimentation and dilution (rainfall and groundwater). This variability was due to the nature of the wastewater but there was a large decrease in the *Giardia* spp. content in all zones during the dry season. Helminth eggs showed little variability within the three zones and there was no obvious difference between the wet

and dry seasons. However, both *Giardia* spp. and helminth eggs were relatively high in zone 3 at the farther reaches of the distribution system, which suggests a possible direct contribution from local discharges, since population in this zone is relatively high (Figure 4). The concentrations of microorganisms reported are similar to those previously described for untreated wastewater in Mexico [19] and are similar to untreated wastewater from other countries [21, 22], but much greater than concentrations seen in effluents of treatment plants [19].

Parameter	Concentration	Description		
Faecal Coliforms,	$6.53 \pm 0.58 \ (5.15 - 7.84)$	Traditional indicator of faecal contamination; may indicate the presence of		
Log(CFU/100 mL)		pathogens; behaves similarly to other bacteria under different		
		environmental conditions.		
Streptococcus faecalis,	5.15 ± 1.0 (2.04-6.20)	Intestinal bacteria found in the faeces of all warm-blooded mammals; useful		
Log(CFU/100 mL)		for indicating the quality of water for recreational use and also for reuse.		
Clostridium perfringens	4.70 ± 0.81 (3.00-5.97)	Anaerobic bacteria that have been recently used as an indicator of faecal		
spores, Log(CFU/100 mL)		contamination; they form a resistant spore commonly found in faeces which		
		is more resistant to disinfection and adverse environmental conditions that		
		many pathogens; the presence of vegetative cells indicates recent		
		contamination while spores imply prior contamination.		
Somatic bacteriophages,	3.63 ± 0.77 (2.41-6.41)	Viruses that infect bacteria; used as indicators based on ease of detection by		
Log (PFU/mL)		analytical laboratories; the coliphage group has been the best model for		
		enteroviruses, given their similar physical structure and their greater		
		resistance to treatment processes (such as chlorination); it is always present		
		and relatively abundant in wastewater; easily detected over a short time		
		period (24 h).		
Giardia spp., Cysts/L	2,231 ± 231	Pathogenic protozoa; single celled organisms that develop in two ways: as		
	(450-10,000)	trophozoites, and as cysts; Infection results from the consumption of a		
		mature cyst that is resistant to the gastric juices; the trophozoites can		
		become cysts again in a process that is apparently aided by unideal luminal		
		conditions, and they are then expelled in the faeces of persons with		
		symptoms of the illness or with no apparent symptoms. Cysts can survive		
		and remain active for weeks, months or even for periods of up to 7 years;		
		capable of forming cysts (resistant structures) under adverse conditions, this		
		organism represents a serious health risk.		
Helminth eggs, eggs/L	13.1±6.2	Group of parasitic and free-living worms of various sizes and shapes; they		
	(1.8-23.0)	cause mechanical deterioration, tissue damage, toxic effects, and blood loss;		
		intestinal parasites cause anaemic malnutrition and/or delayed growth; they		
		present high resistance to chemical and physical conditions and thus are		
		considered the most resistant form of parasites; they have the ability to		
		survive long periods of time in biosolids and soil (up to 6 years after their		
		initial application).		

**Table 1.** Microbiological quality of raw wastewater and characteristics of selected indicators (values in parenthesis indicate range of concentration).

Although it was expected that concentrations of microorganisms would be lower in the wet season due to rainfall dilution, this happens intermittently as the wet season is characterized by irregular intense storms. An additional issue is that the wastewater is mixed with stored rainwater and wastewater depending on the amount of water required by farmers and thus its quality exhibits some variability.

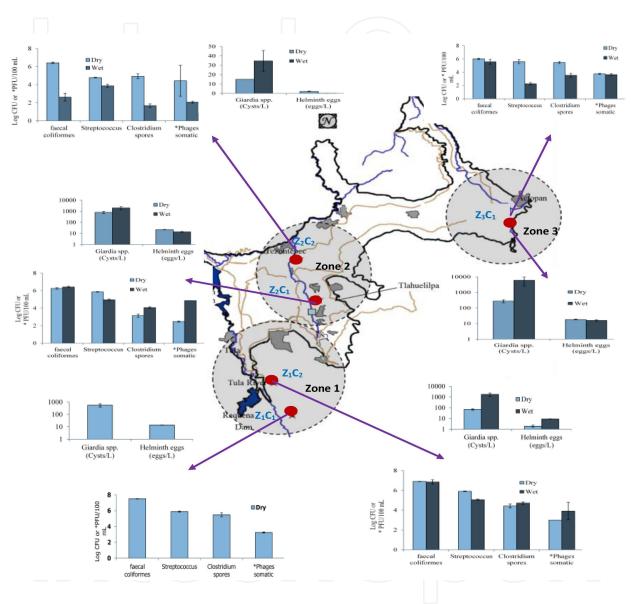


Figure 4. Concentration of selected microorganisms in different zones of the Tula Valley (Adapted from [10] and [23])

#### 4.3.2. Emerging pollutants

Emerging pollutants are defined as those unregulated pollutants that may be controlled by future regulations depending on their potential effects on health and ecosystems [24]. Advances in analytical techniques for their detection and an increased understanding of their effects on public health and the environment, has increased the need to establish discharge limits worldwide. During the last two decades, the study of emerging pollutants (such as pharma-

ceuticals, personal care products, and endocrine disruptors included as additives of gasoline, plastics, or detergents) in the environment has gained scientific attention. Many of these studies have been performed in aquatic ecosystems in the United States of America and Europe [24]. However, for most of the emerging pollutants there are still insufficient data to indicate their risk and ecotoxicity and thus their effects on human health or aquatic organisms remains unknown. Many of these compounds reach the environment through wastewater and they may be removed in wastewater facilities, however, some of them remain in treated effluents and enter the environment, eventually reaching groundwater [25, 26].

Initial reports on pharmaceuticals in wastewater were published in the United States in the 1970's [27], however, even though their presence has been related to fish toxicity, little attention has been given to them [28, 29]. Considering that developed countries have detected emerging pollutants in water and wastewater, even after treatment, their presence in wastewater from the Metropolitan Area of the Valley of Mexico was clearly expected. Furthermore, the fact that many prescription drugs in developed countries are considered over-the-counter medications in Mexico, there is a higher probability of finding their active ingredients in wastewater.

On the other hand, agricultural irrigation favors the transport of pollutants, such as pesticides, from soil to groundwater. Several factors influence their incidence in groundwater, including soil permeability, the depth of the unsaturated zone, geological composition, as well as the solubility, partition ( $K_{o/w}$ ), and dissociation (pKa) values of the pollutant. These conditions may promote their adsorption to soil particles or their leaching to groundwater. It has been reported that wastewater irrigation, treated or untreated, increases the concentration of pesticides in groundwater [30].

As mentioned previously, the presence of emerging pollutants was expected in wastewater from the MAVM, and thus at the Tula Valley, and was confirmed by different studies. Some of them [31] analyzed wastewater samples and found a variety of compounds with the exception of clofibric acid. Concentrations found were similar to those reported for wastewater samples taken at treatment plants (Table 2), but higher than those reported for their effluents [32, 33]. The concentrations of naproxen, a widely use pharmaceutical in Mexico, as well as salicylic acid were higher than those reported in other countries, while ibuprofen, diclofenac, are variable in comparison. Other compounds like 4-nonylphenol and diethylhexyl phthalate were the predominant endocrine disruptors in wastewater. Levels of Triclosan, bisphenol-A, butylbencylphtalate, estrone, and  $17\beta$ -estradiol were similar to those reported elsewhere [33].

It was observed that dilution occurred during the rainy season when reported concentrations of endocrine disruptors were about half of those during the dry season. It should be noted that wastewater at the Tula Valley includes precipitation from the MAVM. Nonetheless, acid pharmaceuticals remain similar in both seasons which may suggest another phenomenon.

Siemens *et al.* [39] concluded that six acid and five basic pharmaceuticals measured in raw wastewater, irrigation water, and spring water, were reduced in concentration along the wastewater distribution canals. However, the authors suggest that acid compounds may permeate clay soils and exhibit poor removal compared to basic or neutral chemicals. Nevertheless, none of the reported compounds were found in spring water.

Compound	Germany <sup>a,c</sup> ng/L	Spain <sup>d</sup> ng/L	Finland <sup>e</sup> ng/L	Brazil <sup>b,c</sup> ng/L	Mexico <sup>f</sup> ng/L
Ibuprofen	3,400	-	13,100	3,300	2,500
Diclofenac	2,000	2,600–5,700	-	800	1,607
Ketoprofen	300		2,000	500	447
Salicylic acid	340-5,400	5			29,867
Naproxen	440	1,800–4,600	4,900	600	13,620

<sup>a</sup>[34, 35] <sup>b</sup> [36]; <sup>c</sup>[25]; <sup>d</sup>[37]; <sup>e</sup>[38]; <sup>f</sup>[31].

**Table 2.** Concentration of pharmaceutical compounds in wastewater.

#### 4.4. Natural attenuation of basic parameters, organic pollutants and pathogens in soil

In the Tula Valley, known as the largest case of indirect wastewater reuse for human consumption in the world, natural soil aquifer treatment (SAT) has been taken place for more than 100 years, recharging the aquifer and acting as a barrier to prevent contaminants from entering it. The local aquifer is being recharged at a rate of 25 m³/s due to the infiltration of untreated wastewater from unlined irrigation channels, storage dams, and excess water used for irrigation (flood irrigation practice; [12]). During percolation, natural soil infiltration occurs through unsaturated soil; as the effluent moves through the soil and the aquifer, it can undergo significant quality improvements through physical, chemical and biological processes.

Non-intentional natural SAT has treatment benefits in the unsaturated zone in the Tula Valley. It acts as a natural filter, and produces groundwater of acceptable quality due to the characteristics of the soil-aquifer structure, the residence time, and the history of the complex geohydrological system. Table 3 showed the historical data of wastewater and aquifer water quality variability obtained from studies performed at the site. It indicates that parameters related with salinity behavior are present in groundwater, in addition to parameters such as nitrate, and solids in dissolved phase. In contrast, the percentage removal of pathogenic organisms and emerging pollutants through unsaturated soil is greater than 50%.

There is evidence that during infiltration the vast majority of microorganisms are retained in the first few centimeters of soil [40], however their potential fate is influenced by size ranges of microorganisms (20-80  $\mu$ m for helminth eggs, 1  $\mu$ m for bacteria, 4-12  $\mu$ m for protozoa), type of soil, and even soil organic matter content for bacteriophages (20 to 200 nm), and therefore for viruses; thus the migration of microorganisms through soil does not always allow complete attenuation.

Parameters	Units	Wastewater (Min- Max)	Aquifer (Min- Max)	% removal (mean)
	Basic ana	lysis		
Total Dissolved Solids, TDS	mg/L	409-1123	546 -1586	-55.5
Alkalinity	mg/L	324-600	256-748	-7.8
Nitrate (as N)	mg/L	0.02-0.47	0.0-32	-8014
	Salinity related	d analysis		
Electrical conductivity	μS/cm	1734-3000	1755 -4187	-18.0
Redox potential	mV	-5137	-79.111.0	-38.7
Sodium	mg/L	56-215	103 -361	-38.3
Potassium	mg/L	19-13190	10.5 -107	98.0
Hardness Ca	mgCaCO <sub>3</sub> /L	90-131	70-787	-218.4
Hardness Mg	mgCaCO <sub>3</sub> /L	12-51	17 -301	-65.7
Total hardness		122-172	114-1006	-181.8
Bicarbonate	mg/L	270-504	212-652	-12.8
Sulfate	mg/L	53-2492	190 -3025	-15.0
	Microbiologica	l analysis		
Total Bacteria count (37 °C)	Log CFU/100 mL	6.4-7.7	1.3-4.2	57.6
Faecal coliforms	Log CFU/100 mL	5.6-7.5	0.0-2.9	70.8
Enterococci	Log CFU/100 mL	2.3-5.9	0.0-1.9	79.3
Clostridium spores	Log CFU/100 mL	3.2-5.6	0.0-2.6	61.1
Somatic Bacteriophages	Log PFU/100 mL	2.5-4.9	0.0-2.8	52.6
Giardia spp.	Cysts/L	70.3-3233	0.0-600	89.4
Helminth egg	Ova/L	1.9-21.9	0.0-2.9	96.0
	Emerging po	llutants		
Clofibric acid	ng/L	< LOD	< LOD-0.39	
Ibuprofen	ng/L	1325-4700	0.05-1.46	99.99
Salicylic acid	ng/L	13580-72979	0.02-27.7	99.98
2,4-D	ng/L	295-2641	< LOD-0.48	99.99
Gemfibrozil	ng/L	640-750	< LOD-0.1	99.99
Naproxen	ng/L	5861-16336	0.04-3.266	100.00
	ng/L	82-500	0.02-0.83	99.89
Diclofenac	ng/L	1240-3424	< LOD-3.75	99.98
4-nonylphenol	ng/L	6970-38130	0.81-67.6	99.93

Parameters	Units	Wastewater (Min- Max)	Aquifer (Min- Max)	% removal (mean)
Pentachlorophenol	ng/L	40-110	< LOD-0.33	99.81
Triclosan	ng/L	360-2880	< LOD-22.6	99.83
Bisphenol-A	ng/L	700-6230	0.02-153	99.54
Butil-bencilphthalate	ng/L	125-2959	0.03-308	97.78
Bis-2 ethyl(hexyl)phthalate	ng/L	4664-70200	3.07-933	99.97
Estrone	ng/L	14-100	< LOD-0.24	99.92
17β-estradiol	ng/L	6.8-22	< LOD-0.06	99.90
17α-etinilestradiol	ng/L	<lod< td=""><td>&lt; LOD-0.05</td><td></td></lod<>	< LOD-0.05	
Carbamazepine	ng/L	200-275	0.14-193	85.89

Adapted from [23, 31, and 39] (ND: Not detected LOD: Lower limit of detection).

Table 3. Water quality variability for wastewater and the Tula Valley aquifer and natural removal percentages (SAT)

Data recorded over many years regarding the attenuation of emerging pollutants through soil show a mean reduction of 86 percent of the chemical load in the wastewater used for irrigation (Table 3). The main attenuation processes such as adsorption and biodegradation have been confirmed in experimental studies with local soil samples, and enriched samples [23]. The most important result of this research is the observation that large amounts of suspended and dissolved organic matter in soils [41], and even in raw wastewater, improve the adsorption of emerging pollutants in soils, and therefore their final low content in groundwater; this verifies previously published results with regard to the role of natural SAT in removing pollutants [26, 42]. In fact, the sorption behavior of three pharmaceuticals (naproxen, carbamazepine, and triclosan), a plasticizer Bis-2 ethyl(hexyl)phthalate, and the surfactant metabolite 4-nonylphenol in wastewater irrigated soil, was analyzed for different soil depths sampled from the Tula Valley, showing that the potential migration of these compounds to the aquifer depends on the physical and chemical characteristics of soil, such as organic matter content, and clay/sand percentages [43, 44].

Other studies have shown that heavy metals have accumulated in soil in the Tula Valley (for example cadmium, nickel, and lead) and their retention is associated with the length of time that wastewater has been used for irrigation [45].

It should be mentioned that a large wastewater treatment plant is currently under construction to treat the MAVM's wastewater and reduce pollutant load to the Tula Valley. This plant, Atotonilco, is designed to treat 35 m³/s with a combined process (23 m³/s with a biological process and 12 m³/s with a physicochemical process) that may partially remove organic matter and the bulk of pathogens. However, the effect on soil attenuation must be evaluated as Gibson *et al.* [41] suggest that a reduction in the amount of organic matter that gets into irrigated soils

may affect sorption processes, which could have an impact on the removal of emerging pollutants, pathogens, and heavy metals.

#### 4.5. Impacts of wastewater irrigation in the Mezquital Valley

Reuse in the Tula Valley has functioned as a discharge route for the closed basin of the MAVM at a relatively low cost. The characteristics of the water, in terms of organic matter and nutrients have significantly increased crop productivity in the Valley and have allowed farmers to harvest up to five crops per year of alfalfa, fodder oats, tomato, barley, and maize. Resulting yields in the Tula Valley are 71 to 150% higher than those obtained with rain-fed agriculture [19]. Thus, the contribution of nutrients represents estimated annual savings of \$180 to \$200 million USD. To exemplify this, agricultural production for 2011-2012 in the Irrigation Districts of the Tula Valley reached \$418 million USD [46].

Since 81% of the main distribution canals as well as 52% of lateral distribution canals are unlined, 80.2 Mm<sup>3</sup>/y of conveyed wastewater infiltrates to the aquifer [47]. It is with this infiltration that wastewater quality is improved before reaching the groundwater through processes like adsorption or biodegradation, which depend on contact time and filtering distance. As an example, travel times from the irrigated fields to the springs are estimated to be 3 to 5 days with groundwater velocities of 0.02 to 6.0 m/d [16].

This infiltration has also incidentally recharged the Tula Valley aquifer for more than 100 years, at a recharge rate estimated at 25 m³/s and equivalent to 13 times the natural recharge [16]. As a consequence, the water table has risen from a depth of 60 meters in the 1950's to 4 meters in the southern part of the Valley and several springs with flows between 40 and 600 L/s have appeared [16]. The amount of available water with a relatively good quality allows some areas to be used to grow vegetables like tomato, lettuce, cabbage, beetroot, cilantro, radish, carrot, spinach, and parsley [16]. At the same time, surface and groundwater produced by wastewater infiltration provide drinking water to approximately 500,000 people after only treatment with chlorine for disinfection [12].

In contrast, wastewater reuse in the Tula Valley has some negative impacts on the local population and the environment. Due to the pathogens and parasites contained in wastewater, the incidence of gastrointestinal diseases has increased by more than 16 times in children living in the irrigation area, compared to children unexposed to wastewater [48]. In addition, microbial and organic pollutants have been detected in the soil matrix of the irrigation areas at various depths [41, 44]. Heavy metals have accumulated in the upper soil layer [49], and Siebe [45] suggests that eventually the retention capacity of the soil might be exceeded with the risk of groundwater pollution. Moreover, the gradual salinization of local soils has caused the loss of more than 2,000 ha of cropland [17].

### 4.6. Conventional wastewater treatment processes, control of micropollutants, and hazardous substances in water

Even though the soil filters a large amount of pollutants from wastewater with removals above 90% [12], some studies have demonstrated that in some areas the filtered water still contains

bacteria (coliforms and enterococci) and viruses [40, 50]. Filtered water presents a high concentration of salts (indicated by sodium, sulfates, carbonates, bicarbonates, chloride, calcium hardness, electrical conductivity, and potassium), and furthermore, total dissolved solids and nitrates exceed the limits set by the Mexican regulatory authorities (1,000 and 10 mg/L, respectively).

At the same time, several studies [23, 31, 40, 41, 43, 44] report the presence of emerging pollutants in springs and wells, which poses a health risk to local population (approximately 500,000 inhabitants) that use the water for human consumption (Figure 5). These pollutants exhibit low removal by conventional treatment processes and thus alternative technologies, such as membrane filtration (nanofiltration or reverse osmosis), have been proposed [51]. Different studies have demonstrated that nanofiltration removes emerging pollutants, including those with low molecular weight, in particular pesticides [52], pharmaceuticals [51], and endocrine disruptors [52].

As a result, to improve the quality of spring water in the Tula Valley, a pilot plant with a nanofiltration membrane was installed in the Cerro Colorado Spring (Figure 5) [53]; The pilot plant (11.4 m³/d) operated for 800 hours and included a pumping system, a prefilter, a softening unit for reducing potential scaling, and a nanofiltration module (Figure 6). The nanofiltration (NF) membrane was selected after laboratory trials on a pilot cell. The pilot plant had the instrumentation required to measure flow, pH, TDS, as well as pressure along the system. Recovery (permeate) was maintained at more than 66 % of the influent.



Figure 5. Cerro Colorado Spring at the Tula Valley.

Nanofiltration demonstrated its effectiveness for removing organic matter (92% of total organic carbon), salinity (60% of total dissolved solids and 75% of electrical conductivity) and 100% for selected microorganisms (faecal coliforms, faecal streptococci, *Salmonella* spp., *Clostridium perfringens*, *Giardia* spp., and bacteriophages). With respect to emerging pollutants, membrane selectivity varied from 5 to 6% for salicylic acid and nonylphenol up to more than 75% for gemfibrozil, butylbenzyl phthalate, carbamazepine, and diclofenac. Treated water (permeate) met the limits for drinking water and thus it could be considered suitable for human consumption.

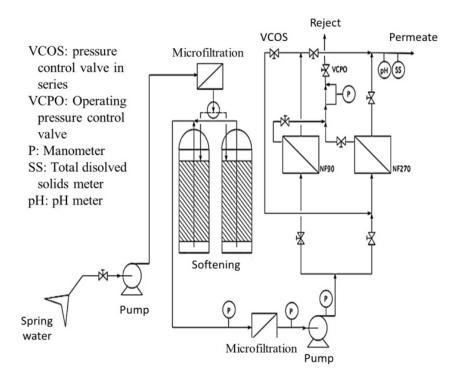


Figure 6. Flow diagram of the nanofiltration pilot plant at the Cerro Colorado Spring, Tula Valley.

Based on such studies, filtered water from the Tula Valley is being considered as a potential source for supplying drinking water for Mexico City and studies have determined that 6.5 m<sup>3</sup>/s may be sourced from local groundwater [54].

#### 5. Forthcoming expectations and recommendations

Findings confirmed by 15 years of research into the complex Tula Valley hydrological and hydraulic system, are:

• Wastewater reuse for irrigation has proven to be an alternative in the water-scarce region of Tula Valley.

- Agricultural irrigation in the Tula Valley using raw wastewater from the Metropolitan Area
  of the Valley of Mexico has been performed for more than 100 years and several studies
  have demonstrated that complex phenomena occur.
- One of these is the natural SAT in the saturated soil zone, which even occurs today after a century of irrigation, mainly in terms of the retention of typical pathogenic organisms and emerging contaminants in wastewater of urban origin.
- The observed level of natural removal through filtration by soil exceeds the expected removal levels from conventional wastewater treatment, not only for emerging contaminants, but also for microorganisms that are difficult to remove by chlorine disinfection.
- Non-intentional aquifer recharge with water purification by natural infiltration, achieved through agricultural reuse of untreated wastewater, has provided new water sources for the local population with the potential to supply water for human consumption to the large city from which comes the reused wastewater originates.
- Advanced treatment studies conducted by our research group have shown that it is possible
  to treat water from aquifer for safe water supply.

There is no doubt about the benefits of wastewater reuse for irrigation, but some potential impacts should be considered:

- Even though the soil matrix retains pollutants, the limit of the soil retention capacity remains uncertain, largely for metals and emerging compounds.
- The content of organic matter in soil has been a key factor in the natural attenuation of water pollution, however, the impact of future changes in the quality of water for irrigation purposes in Tula Valley, may affect removal capacity.
- Soon, the Atotonilco WWTP will supply wastewater to the irrigation canals with a lower content of organic matter and nutrients. These are factors which have previously favored the natural process of the SAT and the development of agricultural activity in the area. These reductions may affect complex physicochemical processes in the future, and may have impacts on soil and groundwater quality that cannot be predicted.
- The use of treated wastewater for irrigation, will allow the growing of crops normally eaten raw, which up till now has been forbidden by national legislation. Other benefits will be the reduction of direct exposure of farmers to pollutants in untreated water.
- Finally, with regard to national and international legislation for irrigation, it is evident that local studies are required for establishing permissible limits. This is because existing guidelines require wastewater treatment levels which are economically onerous, mainly for developing countries, and moreover, do not include limits for other pollutants present in urban wastewater (pharmaceuticals and personal care use products). Wastewater will become more and more attractive for agricultural reuse, given the current and future problems of water scarcity.

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