Physicochemical and Microbiological Quality of Roof-harvested Rainwater in Urban Areas: A Review

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Abstract
This work proposes to consider the quality of roof-collected rainwater. There are three main stages. In the first stage, rainfall washes out the urban atmosphere with contaminants from aerosols and gases. The second stage refers to the catchment, where there is contamination due to the wash-off of particles settled on the roof's surface roofing materials. The third stage refers to the first-flush, storage and plumbing system. In each stage, different processes take place and add specific contaminants to the initial precipitation. Only in the third stage, after the discard of the high-polluted initial rainwater, some physical processes (for example sedimentation and correction of pH) can also improve the quality of the rainwater harvest. This approach offers a clear view of the overall contamination processes that take place in a rainwater harvesting system.The most common microbiological and physicochemical contaminants that can be found in rainwater harvesting system were considered, together with the eventual presence of waterborne pathogens and emerging chemical contaminants, according to an extensive review of previous scientific works.
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1.0 Introduction

The growth of urban areas demands a continuous increase in the amount of supplied fresh water. As most urban springs and aquifers are overexploited or polluted, water must be imported from distant reservoirs with the corresponding energy cost and losses due to leakages. Conflicts regarding the use of water resources have arisen in many regions of the world. (DeJong et al., 2013).

Therefore, any technology that can produce some amount of water with an acceptable quality, is more than welcome. Together with water conservation programs, the interest for water sources that are alternative to traditional water supply systems has increased in recent times. Among these alternatives are; water reuse, water reclamation and rainwater harvesting. Therefore, water harvesting that was only a valid resource in rural areas with no water mains coverage, is now considered as a convenient option for urban and developed areas.

Rainwater presents a series of advantages regarding some of its physicochemical parameters, when compared with water from the mains or groundwater. It has lower hardness and total suspended solids. Well designed rainwater harvesting systems with clean catchments, and storage tanks, as well as simple treatment can offer water for non-potable uses with very low health risk. Common available treatments for harvested rainwater are: the application of settling tanks, disinfection combined with membrane filtration, reverse osmosis (Wang et al., 2014), heat treatment, solar disinfection (Ahammed et al., 2014) and slow sand filtration followed by chlorination (Moreira Neto et al., 2012). In addition, point-of-use systems such as UV lamps and ozone disinfection (Lopez, 2014) are a growing trend in rainwater treatment for drinking purposes. The improvement of roof-harvested rainwater to reach potable quality standards is gaining popularity in urban environments, while in rural sparse areas where water mains is not available this option may greatly increase the local sanitary conditions.

Given this context, there is need for information and guidelines about the common sources of pollutants or contaminants of roof-harvested rainwater in urban areas. The literature on this issue is wide and it assesses the following connected issues: the detection of specific chemical or microbiological contaminants in rainwater; comparative rainwater analysis; the deposition of atmospheric pollutants in urban surfaces, their build-up and wash off; and the influence of roof and storage material, among others.

This review aims to summarize and analyze relevant scientific works that have addressed some parts of this problem, compiling them to understand the mechanisms and pollution sources that influence the final quality of roof-collected rainwater in urban areas. Chemical contaminants, together with the most common bacterial and waterborne pathogens was considered (Kwaadsteniet et al., 2013). This review takes into account three stages that would affect the quality of the rainwater and their different pollution loads. Stage one refers to the collecting of atmospheric pollutants by the rainfall (wet depositions). Second stage is the wash-off of particles settled on the surfaces (dry depositions). Third stage is collection, pre-tank filtration and storage in the rainwater tanks.

2.0 Quality of roof-harvested rainwater

2.1 Rainwater contamination due to atmospheric deposition in urban environments

2.1.1 Atmospheric deposition processes

Atmospheric deposition is the transfer of atmospheric pollutants (dust and particulate) to terrestrial and aquatic ecosystems (Amodio et al., 2014). Many of these pollutants may be present in urban environments, at different rates according to the intensity of road traffic and the proximity of industrial areas. Sources of contaminants deposited from the atmosphere may be road traffic (Fang and Zheng, 2014), sea spray, industrial and rural activities and local dust.

Their presence in the atmosphere of the towns and subsequently in the rainwater that flows from the roofs can have a great spatial variability. It also depends on meteorological factors (wind velocity, temperature, relative humidity), particle characteristics (size and shape).

Dry deposition is a process that is not influenced by precipitation: particles are carried out by the wind and eventually fall from the atmosphere into the urban soils and surfaces.

Occult deposition consists of water droplets deposited by the interception of fog, mist, or clouds, which play a significant role only in the case of frequently cloud-covered zone, but are negligible in most of the urban areas. Wet deposition refers to the rain washout of the air, which captures the air pollutants inside the rain drops and transfers them to the soil.

2.1.2 Nitrogen

Organic nitrogen in the form of pollen, spores, bacteria, and other substances can also make a significant contribution to the total nitrogen concentration in the rainfall, depending on the season of the year and on wind and climatic conditions.

The presence of nitrate originated from anthropogenic activity in the rainfall is recurrent in urban areas worldwide. Most of the nitrate and other nitrogen oxides as well as ammonium present in the stormwater runoff may end up contributing to the pollution of groundwater in urban areas (Umezawa et al., 2009).

2.1.3 Phosphorus

Together with the loads of nitrogen due to wet and dry depositions, ammonium and phosphorus from natural sources such as bird feces and lichens that contaminate the roof runoff can also be added to the rainwater. In addition to the organic phosphorous that would be later added to the runoff, the concentration of inorganic phosphorous present in the rainwater (in the form of phosphate, $PO^3{}_4$) can by itself exceed the concentration threshold for algal blooms of 0.03 mg P/L.

2.1.4 Other ions

Rainfall contains several ions from the atmosphere or transported within the rain fronts. These ions can originate from various pollutant sources, both natural or from anthropogenic activity.

Excess material from the soil can increase concentration levels of sodium (Na^+) and potassium (K^+) ions, while calcium (Ca^{2+}) can be associated with the occurrence of dust storms. Most of the sulfate ($SO^2{}_4$) present in the rainwater that falls over the ocean and coastal areas originates from sea spray.

Coastal rainwater is strongly influenced by the sea and thus major ions derived from sea-salt aerosol can make a significant contribution to the total ions and trace metal content of coastal rainfall (Halstead et al., 2000). Sea salt aerosol contains both inorganic salts and organic matter from the ocean. It is mainly constituted of sodium chloride (NaCl) together with other ions which are common in sea water: K^+, Mg^{2+}, Ca^{2+} and $SO^2{}_4$.

Sazakli et al. (2007) monitored rainwater quality in a Greek island for over three years, concluding that microbiological parameters were affected mainly by the cleanness level of catchment areas, while chemical parameters were influenced by the sea proximity and human activities. Some ions found in rainfall were from sea salt origin: ions chloride, sodium, and magnesium (Mg^{2+}). This study also identified the ions originated in a pollutant source that acidifies rainwater: nitrate and sulfate from road traffic and industries, ammonium ($NH^+{}_4$), and hydrogen (H^+).

In contrast to the above studies that address coastal rainwater, chemical characteristics of rainfall and wet deposition in continental, sea-distant regions were analyzed by Yang et al. (2012) in Beijing and Robakidze et al. (2013) in the Urals (Russia). Aikawa et al. (2014) studied the influence of site in the atmospheric deposition of NO_3 and $SO^2{}_4$ in rainwater. The results showed that in urban sites the contribution of washout in NO_3 depositions was 70% while in suburban and rural areas this contribution was 66%. Washout accounted for 50% of the $SO^2{}_4$ depositions in suburban and rural sites and 80% in urban sites.

2.1.5 Acidity

Acidity in rainwater is highly dependent on the trajectory followed by the air mass and can be influenced by the passage over distant industrial areas. Rainfall acidification can originate from anthropogenic activity. Nitrogen oxide gas (NO_x) and sulfur oxide gas (SO_2) are exhausted from chimneys of factories and from automobiles. Later on, photochemical reactions transform these compounds into nitric acid (HNO_3) and sulfuric acid (H_2SO_4).

The atmospheric deposition of organic acids may also be significant in some urban locations, and can also influence the pH of the rainwater. The presence of carboxylic acids and aldehydes (which can oxidate and thus become a relevant source of organic acids) in rainwater was addressed by Peña et al. (2002). In this study, the most frequent acids found in the rain samples were formic and acetic acids. The major aldehydes present were acrolein, formaldehyde and acetaldehyde. Although precipitation in the area of study was influenced by the presence of a thermal plant, it was established the prevalence of natural vegetative origins for both carboxylic acids and aldehydes versus anthropogenic emissions.

2.1.6 Heavy metals

Atmospheric depositions of heavy metals are among the most important pollution loads of urban surfaces. Cizmecioglu and Muezzinoglu (2008) studied the solubility of deposited airborne heavy metals and expressed a bigger concern regarding the ecotoxic properties of heavy metals in wet deposition far more than dry deposition. This due to the high soluble fractions of the dry deposited heavy metals. The trace metals are bounded into the particles settled on the urban surfaces due to dry deposition. The release of these metals from the particles to the runoff water occurs within a few minutes. The solubility of non-crust-dominated trace metals, such as Pb, can vary with the pH of the rainwater and even during an individual rain event. Metals like Cd, V, Cu and Zn show a higher average solubility than Ni, Cr, and Pb, both in wet and in dry depositions. According to Gunawardena et al. (2013) Zn is associated with relatively larger particle size fractions (>10 µm) and tend to appear as dry deposition, whereas Pb, Cd, Ni and Cu are associated with relatively smaller particle size fractions (<10 µm) and more suitable to wet deposition. The transferring of mercury from ambient air to rainwater in an industrial urban area was addressed by Ghadaksaz zadeh et al. (2014). The values of mercury concentration in rainwater provided in this work, for different urban areas of the world, range from 0.007 to 0.77 µg/L.

2.2 Rainwater contamination due to the wash-off of particles settled on the roof's surface

The best quality rainwater available in urban areas is the one that is harvested after the first roof interception, thus minimizing the effect of rain and the contact with settled particles. After the initial interception of rainwater, the pollution process of rainwater keeps going on as the particles settled on urban surfaces are carried out by the stormwater runoff, and can drastically reduce the initial quality of rainwater. As the leaching of particles occur in the initial stages of a rain event, by discarding the first-flush of the roof's runoff it is possible to drastically increase the quality of the harvested rainwater.

2.2.1 Bacteria and pathogens

Airborne microorganisms and aerosolized bacteria can be transferred to harvested rainwater through atmospheric fallout of coarse particles (dry deposition) with the subsequent wash-off of the catchment's surface and through rainfall (wet deposition). Several microscopic organisms can be usually found on the catchment surfaces, either in the deposition layers or associated to decomposing organic matter. Fresh rainwater falling into the roofs already presents microbial pathogens, even before the interception in the roof's surface and thus provides an incoming bacterial load.

Lye (2002) stated that fecal coliform measurements may prove to be an inappropriate indicator for determining microbial risks associated with the consumption of harvested rainwater. (Kaushik et al., 2014) assessed the microbial quality of rainwater using the following as microbial indicators: E. coli, total coliforms, and enterococci along with total heterotrophic plate counts (HPC). This study obtained levels of E. coli in rainwater within the range of 0–75 CFU/100 mL for a highly urbanized site with tropical climate (Singapore), concluding that despite the presence of microbial pathogens, rainwater harvesting in large water catchment areas is a promising freshwater resource following treatment.

Waterborne pathogens may be present in urban roofs due to biological activity associated with depositions of windblown dirt, fecal droppings from birds and other animals, insects and litter, lichens and mosses, fungus or fallen vegetable material from the surroundings trees (leaves, seeds, flowers), even in clean metallic catchments.

Man-van der Vliet (2014) summarized the concentration values of a series of waterborne pathogens (bacterial, viral or protozoan pathogens) in feces, wastewater, surface water, stormwater and runoff rainwater, from the guidelines for drinking water quality of the WHO.

The results shown on **Table 1** point out to a significant loss of quality in the rainwater from the roofs and having been washed (runoff), reaches the urban soils and flows through the urban drainage systems (stormwater).

Table 1: Concentration of indicator bacteria and pathogens in wastewater, surface water, storm water and runoff *(Man-van der Vliet, 2014)*.

	Wastewater (No/L)	Surface water (No/L)	Stormwater (No/L)	Runoff (No/L)
Indicator bacteria: E. Coli	10^8	10^3	10^3	10^2
Campylobacter	10^6	10^3	10	10
Cryptosporidium	10^2	10	1	1
Giardia	10^2	50	5	1

There was a statistically significant variation in the physicochemical and microbiological quality of the samples depending on the location of sampling. However, most of the samples exceeded the maximum accepted faecal count.

In addition, this study also detected the presence of unacceptable levels of lead in the runoff in about 13% of the samples. The authors concluded that the high prevalence of lead levels is likely to be the result of corrosion of galvanized iron, lead and lead-based paint on the catchment surface rather than atmospheric deposition. Ahmed et al. (2012) suggested that sources of E. coli in rainwater tanks in Australia are likely to be bird and possum feces.

2.2.2 Metals in urban roof catchments

In addition to the biological risk, which can be minimized by periodic cleaning and maintenance, urban roof catchments can also be a source of inorganic contaminants for rainwater.

Rainwater pollution with metals such zinc, copper and some lead is often associated with corrosion of roofing materials. As rainwater is slightly acidic and very low in dissolved minerals, it can dissolve metals and other impurities from materials of the catchment and storage tank, causing discoloration together with bad taste and odor in the harvested rainwater.

2.2.3 Influence of the roofing material

Farreny et al. (2011) provide criteria for the roof selection in order to maximize the availability and quality of rainwater, through a combined approach that studies the runoff coefficient (quantity of harvested rainwater) as well as physicochemical parameters in the runoff (quality). These criteria are related to the roof's slope and roughness. In this study, two roof types are monitored: one made of clay tiles, metal sheet and polycarbonate plastic and one flat gravel roof, concluding that runoff water quality is better in sloping smooth catchments than in flat rough roofs and also that the necessary first-flush volume is highest in flat rough roofs.

Different roof types have an influence on the rainwater quality with zinc concentrations higher in galvanized iron roof catchments while pH, conductivity and turbidity levels are higher in concrete tile roof catchments. Their results also show higher values of nitrates and pH in roof-harvested rainwater in rural areas and presence of lead in rainwater collected in industrial areas and, to a lesser extent, in urban areas. Both rainfall intensity and the number of dry days that preceding a rainfall event significantly influences the quality of run-off water from the catchment systems.

Lee et al. (2012) studied the influence of the following roofing materials: wooden shingle tiles, concrete tiles, clay tiles, and galvanized steel (zinc alloy coated steel). The best rainwater quality was obtained with the galvanized steel roof, where the combined effect of ultraviolet light and the high temperature effectively disinfected the harvested rainwater, whose quality parameters meet the South Korean guidelines for drinking water.

Values from Table 2 illustrate that the initial runoff leaches the greatest part of contaminant deposits from the roof and subsequently corroborate the effectiveness of first flush deviation in the improving of rainwater quality.

Table 2

Average quality values and concentration of contaminants in the roof-collected rainwater, from different roof materials and rainwater. *(Lee et al. (2012)*

		Concrete tile	Clay Tile	Galvanized steel	Galvanized aluminum
pH	Rain water	7.3	7.1	6.1	6.1
	First-flush tank	7.1	7.0	6.4	6.3
TSS (mg/L)	Rain water	45	42	15	24
	First-flush tank	301	210	280	103
Nitrate (mg/L)	Rain water	0.3	1.0	0.02	0.4
	First-flush tank	2.3	2.7	2.7	1.0
T.C. (CFU/100 mL)	Rain water	12	2	Nil	145
	First-flush tank	191	72	70	290
Al (µg/L)	Rain water	99	36	33	135
	First-flush tank	530	240	612	645
Fe (µg/L)	Rain water	48	24	27	140
	First-flush tank	153	152	300	580
Cu (µg/L)	Rain water	20	12	22	64
	First-flush tank	52	33	60	66
Pb (µg/L)	Rain water	5	3	3	0.4
	First-flush tank	12	10	12	2.3
Zn (µg/L)	Rain water	38	19	74	198
	First-flush tank	189	130	420	370

Table 3:
Average quality values and concentration of contaminants in the roof-collected rainwater, from different roof materials. (*Olaoye and Olaniyan (2012*)

	Galvanized roof	Asbestos cement	Aluminum roof	Concrete flat roof
pH	6.1	6.6	6.9	6.1
Nitrate mg/L	0.4	35.5	8.3	9.5
Cu µg/L	<25	4.10 2	2 2.10	2 2.10
Zn µg/L	5700	6102	6102	6102

Table 3 provides values of pollutants found in the first-flush of other types of roofing materials.

2.3 First flush deviation and rainwater contamination during storage
The adequate volume of first-flush that needs to be discarded varies with the intensity of the rainfall event. The first-flush volume also depends on the dry days preceding the rain event. This study reported that, for California's climate, the concentration of contaminants in the first part of the wet season ranges from 1.2 to 20 times higher than the end of the season, as during the dry season a long period for pollutant build-up on the catchment's surfaces is created. Discussion

3.0 Discussion
Before the interception on the catchment surfaces, rainwater can already present some contamination as it has passed through the first stage that influences its quality. The first stage in the contamination of rainwater occurs when rainfall washes out and collects aerosols, gases and thin volatile particles from the urban atmosphere. The source of these contaminants can be local road traffic and industrial activities or be situated far away, depending on the particle characteristics. For the pH of rainfall in urban areas, the values reported from the different studies range from 4.5 to 10.4 and show that anthropogenic activity can have a great effect in this parameter. The presence of other toxic compounds in the rainfall, collected from the atmosphere, can be of particular concern in specific areas. This is the case of some emerging contaminants, such as polycyclic aromatic hydrocarbons (PAHs), perfluorinated compounds, dioxins or carbonyl compounds.

Fecal bacteria washed off from the roof are a common pollutant load and their detection appears in all the reviewed rainwater quality analysis. However, it may not be an adequate indicator of the presence of waterborne pathogens. Bird feces deserve special attention as some of the reviewed studies have linked them to the presence of Salmonella in rainwater. Concentration values of indicator bacteria and pathogens in feces, wastewater, surface water, storm water and runoff were presented in Table 1.

Considering physicochemical contamination during this stage, it can be significant the presence of heavy metals in the particle matter settled on the catchment surfaces, through wet or dry deposition depending on their solubility. Sea spray was found to be an important source of wet deposition in coastal areas. It was also found that roofing and gutter material can be a source of chemical compounds, with many studies illustrating the leaching of metals as in the case of lead-based paints.

The initial roof runoff contains the highest particulate matter concentrations and with them most of the pollutants, and thus an adequate volume of this first flush should be discarded or deviated. The optimization of the first-flush volume according to the intensity of rainfall and the previous dry period is critical to reach an effective removal of rainwater pollutants that would otherwise enter the rainwater tank.

The effectiveness of first flush deviation in the improving of rainwater quality is has been studied (Lee et al., 2012; Mendez et al., 2011). In Table 2 results from these studies were presented, comparing the average quality values in the first-flush tank and in the rainwater tank, for different roof materials.

In Table 3, (Olaoye and Olaniyan, 2012) also presented average quality values and concentration of contaminants in first-flush samples of roof-collected rainwater, considering different roof materials. It was found that the type and material of the catchment's surface can influence the contamination of the runoff in a significant manner.

During the storage stage, an increase in pH and sedimentation are two physical phenomena that contribute to the enhancement of the quality of the rainwater harvest. The leaching of chemical and organic compounds should not have a considerable effect on the quality of the stored rainwater if a good maintenance and a good selection of the waterproofing paint of the tank is done.

4.0 Conclusions
The proposed approach for the study of this topic combines the assessment of both the main physicochemical and microbiological contaminants, together with the physical phenomena behind the scavenging and transportation of these contaminants, and with the practical experiences and case studies about rainwater

harvesting systems around the world that were found in the scientific literature. Therefore, the quality of roof-collected rainwater is considered as the sum of three main stages. In each stage, different processes take place and add specific contaminants to the initial precipitation. Only in the third stage, after the discard of the high-polluted initial rainwater, some physical processes (sedimentation and pH increase) can also improve the quality of the rainwater harvest.

This approach revealed to be useful, as the physical phenomena involved (build-up, scavenging, wash-off) is common for most of the microbiological and the chemical contaminants. It also offers a clear view of the overall contamination processes that take place in a rainwater harvesting system.

Several studies that show rainfall analysis in urban areas were reviewed. Their results illustrate that heavy urbanized areas can present significant concentrations of ions in rainfall such as ammonium ($NH^+{}_4$) as well as nitrate (NO_3) and sulfate ($SO^2{}_4$) from traffic and other anthropogenic activities.

The wash-off of the catchment's surface is the main source of microbiological contamination of roof-harvested rainwater, as airborne pathogens and organic matter are deposited on it.

For the storage stage, the main conclusions are the influence of the correct regulation of the first-flush deviation and the need to follow good practices with respect to cleaning, disinfection and maintenance of the catchment and storage system. Lack of maintenance, poor design and inadequate disinfection regimes are the main threat to rainwater quality during this stage.

A common conclusion found in the different studies addressing this topic is that harvested rainwater needs to be treated before use for drinking purposes. In general, rainwater analysis in the reviewed studies point out to acceptable values of physicochemical parameters. The values obtained for the microbiological parameters indicate a low water quality and subsequently a high health risk. In order to meet the standards required for drinking purposes, point-of-use systems such as UV lamps or ozone disinfection are recommended.

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