Removal of Emerging Contaminants: The Next Water Revolution

E. McBean†*

† School of Engineering, University of Guelph, Guelph, Ontario N1G 2W1, Canada

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ABSTRACT. Thousands of new, emerging chemicals are produced each year, making thorough investigations infeasible regarding their potential detrimental dimensions. As an important step for estimating whether a chemical will result in an exposure pathway and therefore create the potential for a detrimental impact, a coefficient-based strategy consisting of eight key coefficients, is proposed. The strategy is based upon key factors which are used to assess the potential for a chemical to attenuate or change its phase or medium, as part of its fate and transport pathway. The eight key coefficients are described, knowledge of which will assist in determining whether a chemical will result in a fate and exposure pathway change and/or attenuate, as a means of developing a strategy to assess the risks of emerging contaminants. The need for attention to this next water revolution to develop a strategy to assess some of the risks of emerging contaminants is already upon us.

Keywords: emerging contaminants; fate and transport; PCBs; attenuation; octanol partitioning; adsorption; biodegradation; volatilization

1. Introduction

Water is renewable but also finite in availability, and yet, it is a prerequisite for life (Postel, 2000; Lee et al., 2009). The result is that many challenges exist in terms of satisfying the competitive demand for water while simultaneously ensuring the water that is available is of a quality needed for alternative uses, or ‘safe’. Over time, as the depth of awareness surrounding water issues has evolved, so too has humans’ understanding of how to adapt in response to these newfound problems. In this regard, society is again at a ‘turning point’ where humans are faced with both the need for new knowledge and awareness that significant effort is required; significant effort is now needed to identify which emerging contaminants are of greatest concern. This paper describes the evolutionary pathways that humans have historically adopted governing water availability and access, and then provides some methodologies that are useful for purposes of dealing with emerging contaminants, to address issues that we are only now starting to understand.

2. Historical Evolution of Water Use and Response

The historical response to water problems concerning characteristics/availability is informative to better appreciate which has occurred over many centuries. These advancements are both the solutions adopted, and the improved understanding specific to their underlying circumstances:

(i) Hunters and Gatherers – Between 25,000 and 16,000 years ago, with extremely dry and arid conditions, small, roving bands of 25 ~ 50 people existed through hunting and gathering. Seasonal scarcities forced these bands to nomadically gather by locations neither far from water, nor too close (insects drawn to the humidity).

(ii) Agrarian Societies – Between 16,000 and 8,000 years ago, there was a trend toward sedentary settlements, which came with it an increase in population densities. Clearly, there were advantages to having a number of women, men and children available to harvest the crops. People started to domesticate livestock and hence needed to provide water for this purpose. Settlements were typically located close to springs and streams, providing access to clean water.

An economic revolution for early state societies resulted from the invention of agriculture. Neighbouring villages began to depend on each other for trade. Emerging problems related to early increases in population density were many, including sanitation and hygiene issues, local food shortages, and disease. Farmers sometimes faced water shortages when rivers dried up or changed course and they responded by digging divergent canals, rerouting waterways, and constructing earthen dams.

(iii) Assyrians in Nineveh – About 7,000 years ago, King Sennacherib created a new capital in Nineveh (present-day northern Iraq), an area with little rainfall. Since the King thought it too barren and wanted greenera, a channel was constructed to carry water from nearby mountain streams (70 km from the city) (AAT, 2012; Taggart, 2018). The route had to cross a small valley requiring construction of an aqueduct nearly three football fields in length and as wide as a four-lane highway. The aqueduct allowed for water conveyance to con-
tinue across the depression. Materials for this structure included 2 million carved limestone blocks, each weighing roughly a quarter of a tonne. Nineveh relied on this imported water and was once the largest city in the world with this first structure carrying water to the people.

(iv) *Egyptian Times* – About 6,000 years ago, agriculturalists moved into river valleys of the Nile. The climate was arid but the soil was excellent for crops (if only water could be brought to it). Dams were built along the Nile, and channels were constructed to divert water from the Nile to agricultural fields. This system realized the capability to sustain an increasingly dense population; the population density surrounding the Nile was unprecedented.

(v) *First Recognition of Need for Treatment, to Improve Water Quality* – About 4,000 years ago, ancient Greek and Sanskrit writings indicate people knew heating water could purify it. They also knew to avoid stagnant and marshy water. Use of senses (taste, smell, appearance and touch [temperature]) were employed. As well, about 3,500 years ago, Egyptians identified coagulants (alum) could be used to settle suspended particles.

(vi) *Persian Use of Qanats* – Approximately 3,000 years ago, Persians delivered water using ‘qanats’, a subterranean system of tunnels connecting wells (Motiee et al., 2006). Long distance transport (50 km) of groundwater was accomplished from mountains to low-lying farm land. This allowed farming to extend to marginal desert areas, providing reliable supplies of water for human settlements. The qanat technology spread, starting about 800 BC, to Iraq, Pakistan, Arabia, Egypt, China and others (Globlot, 1979; Baboli and Khaniki, 2005; Motiee et al., 2006).

(vii) *First Urban Sanitation System* – 3,000 years ago, the Indus Valley was advanced in wastewater management. Even as early as 2,500 BC, Harappa and Mohenjo-Daro included the world’s first urban sanitation systems. Houses were connected to drainage channels and wastewater was not permitted to flow directly to the street sewers without first undergoing a degree of treatment. Wastewater was passed through tapered terracotta pipes into a small sump. Solids settled and accumulated in the sump, while the liquids overflowed into drainage channels in the street when the sump was ~75% full. The drainage channels were covered by bricks and cut stones. This, most likely, was the first attempt at treatment at the household level (Lofrano and Brown, 2010).

(viii) *Roman Times* – 2,500 years ago, Rome had a population of ~500,000. Rome needed large quantities of clean water for drinking, domestic use, baths and latrines. The first aqueducts (in Latin, aqua means ‘water’ and duerere means ‘to lead’) were powered by gravity. Water was supplied to agricultural estates (the core of Rome’s economy and wealth). As well, 2,500 years ago, Hippocrates (Greek) invented the first cloth bag filter to remove turbidity. The Romans also built drains and sewers to take away their wastewater, including the world’s earliest sewage system (Cloaca Maxima) (Cosgrove, 1909). After water was used for cooking and bathing, it was then flushed through the sewers, along with human excreta. However, about 2,000 years ago, economic inflation in Rome reached 13% and lasted for 100 years. During that time, the price of corn rose by a multiple of 300,000. Conditions reached the point where it was impossible to levy taxes, resulting in public works (such as sewer maintenance) being neglected. Subsequently, the outbreak of Cloaca Maxima filled with sediments, the Pontine Marshes were flooded, and mosquitoes bred throughout the City. The resulting outbreak of malaria left the population of Rome decimated, the barbarians invaded and the dark ages commenced.

(ix) *The Dark Ages* – From 500 ~ 1,500 AD, the understanding of water issues lost much of its sophistication and advancements in water technology ended. With no scientific innovations and experiments, it was not until the 1,600s (AD) that experimentation with water treatment and interest in clean water technology resumed.

(x) *Widespread Use of Filters* – About 300 years ago, widespread use of filters became common (using wool, sponge, and charcoal), the first water treatment plant was constructed in 1804, in Paisley, Scotland, using slow sand filtration (see Baker and Taras, 1981; Mays, 2013). Since in those days there were not water distribution pipes available, horses and carts were used to distribute the treated water to individual homes.

(xii) *Realization that Miasma Didn’t Cause Illness* – 150 years ago, there was realization that waterborne pathogens were the cause of cholera, and not miasma (or bad air) (Morris, 2008). Dr. John Snow correctly identified that the cause of the cholera outbreak in London was from a drinking well water that had been contaminated by raw sewage. This revolutionized conventional wisdom and finally, successfully demonstrated by a very stubborn Dr. Snow, that good taste and odorless water were not sufficient to ensure water was ‘safe’. Dr. Snow was eventually able to overcome the prevailing thinking of the times. In light of this discovery, chlorine was applied to purify the water. This form of water treatment has had the greatest overall impact on human health in all of history (Morris, 2008).

3. Emerging Contaminants, the Third Water Revolution

The evolutionary pathway described above, to improve access to ‘safe’ water, was both arduous and necessary. Recognition followed that with increasing populations, there was need for efficient water use, and efforts were extended based upon knowledge, toward availability of water appropriate for human consumption and for the environment. This same approach of increasing attention to the need to strive to protect the quality of water has continued over the most recent 100 years. Specifically, recent revolutions in the assessment of water have been occurring, namely:

(i) Revolution 1: Physical and Biological Treatment – these undertakings began ~1930;

(ii) Revolution 2: Nutrient Removal (N&P) – these undertakings began ~1970;

(iii) Revolution 3: Emerging Contaminants – removal efforts are underway but proving to be very challenging.

The term ‘emerging contaminants’ refers to chemicals and
Control of exposure pathways to limit exposure of humans and the environment as a mitigation technique is frequently infeasible. Thousands of routes of exposure exist including, for example, through human excreta (urine) where up to 90 percent of oral medications pass through the body unchanged, adding to the pathways migration by which human exposure to pharmaceuticals may occur (Storteboom et al., 2010; Science Daily, 2019). Some people dispose of unused medications in the toilet, representing another pathway of pharmaceuticals if the wastewater treatment plant is not effective at destruction of the pharmaceutical (as occurs for many pharmaceuticals). Inhalation pathways, dermal pathways, food ingestion, and many more exist.

Recent trends of shorter inpatient care result in sending patients home with their pharmaceutical medicines. This is resulting in pharmaceuticals now being commonly found in wastewater from residential areas (e.g. see McBean et al., 2018b, c). This finding indicates that we cannot restrict our focus of managing pharmaceutical contamination of wastewater to only hospitals as a means of point source control.

Exposure to veterinary medication is increasing with growing consumption/excretion/land application. US livestock and poultry herds require 3.5 times more antibiotics than the total US population (Meat Myth Crushers, 2018). This has caused concern with the magnitude of antibiotics and ‘sedatives’ (tranquilizers and beta-blockers used to relieve anxiety and stress of food-producing animals and hypnotic sedatives are now commonly injected to calm the animals as they are transported to the slaughterhouse (McBean et al., 2018a). The uses in animal agriculture have resulted in the potential for increasing levels of tranquilizers and beta-blockers now being found in receiving waterbodies.

The above indicate the importance of understanding the fate and transport characteristics of specific emerging contaminants, information which will be valuable to establish the pathways which will govern how human and environmental receptors may be exposed.

4. An Approach to Targeting the Key Coefficients to Improve Information Assembly for the Third Water Revolution

With thousands of new chemicals being introduced into society each year, keeping up with detailed knowledge of the chemicals, including dimensions of impact that may occur to humans and/or the environment, is infeasible. Improvements in analytical chemistry (e.g., using liquid chromatography-tandem mass spectrometry (LC-MS-MS)) have been instrumental in facilitating attention to emerging contaminants since the monitoring capabilities now exist, all of which have been instrumental in facilitating the third revolution to be in process. Nevertheless, while the opportunity exists to do the monitoring, the opportunity for the investigation of emerging contaminants is challenged due to the large number of chemicals entering the marketplace.

While destruction of some emerging contaminants in a
wastewater treatment plant is feasible, removal of emerging contaminants by this means requires large investments of both capital and electricity to operate, and frequently, a sequence of unit treatment processes. As well, while destruction of emerging contaminants at wastewater treatment plants in some circumstances is feasible, sampling to determine rate coefficients of destruction of emerging contaminants is typically profoundly difficult (e.g. see Smyth et al., 2007a, b).

Given that thorough investigations will not be feasible due to the large array of emerging contaminants that already exist and/or will soon become present, an alternative strategy can involve the characterization of the potential for an emerging contaminant to create an exposure pathway (e.g., the degree to which it cannot be fully removed/destroyed in a wastewater treatment plant), by identifying the potential to estimate the attenuation of an emerging contaminant, relying on ‘key coefficient’ properties. This can be accomplished by improving our understanding of the fate and transport of other chemicals with similar characteristics with the same property. To demonstrate, consider the list of possible key properties is provided in Table 2 which can provide a guide to whether a chemical will attenuate by specific mechanisms.

It is acknowledged that the list of eight key coefficients listed in Table 2 is not necessarily exhaustive; others may be identified at a later date. Regardless, the merit of the methodology to facilitate systematic thinking is the primary objective, although the eight selected are expected to be the fundamental underpinning as they are well known, relatively widely available, and key to having a methodology that will provide the needed insights to commence the process of prioritizing efforts to control emerging contaminants.

As well as the above information of key properties, guidance on whether a particular chemical is a known or probable carcinogen can be found on US EPA, IARC and NTP websites (US EPA, 2018; International Agency for Research on Cancer (IARC) 2018, and NTP (2018); see also, for example, McBean (2019) for guidance on carcinogenic classification mechanisms and source information).

5. Pertinent Characteristics of Chemicals

Specific chemical characteristics describe the phase or medium in which a chemical is likely to be found. For example, Henry’s Coefficient describes the volatility of a chemical and hence indicates the potential for a chemical to volatilize and be present in gaseous emissions. Hence, the strategy involves use of these fundamental chemical and biological characteristics to project whether the individual chemicals would, for example, tend to volatilize and become an air emission as a pathway of exposure.

This accomplishes two fundamental aspects: the fate aspect of the chemical (e.g., will the chemical biodegrade or combust in a flare?), and secondly, will the chemical change media (e.g., move from the aqueous domain by volatilizing to the gaseous domain) which can be used to assess whether air emissions are the primary pathway. The key coefficients recommended for use are as follows:

(i) Octanol/Water Partition Coefficient

The Octanol/Water Partition Coefficient is a unitless coefficient defined as:

\[ K_{ow} = \frac{\text{Concentration in octanol phase}}{\text{Concentration in aqueous phase}} \] (1)

The effect of temperature on \( K_{ow} \) is not significant. The value of \( K_{ow} \) represents the tendency of a chemical to partition between the organic phase and the aqueous (leachate) phase where:

If \( \log K_{ow} < 1 \), the chemical is hydrophilic or tends to the
water phase; and

If \( \log K_{ow} > 4 \), the chemical is very hydrophobic, indicating the chemical tends to the octanol phase.

(ii) Adsorption Coefficient

The extent to which an organic chemical is partitioned between the solid and solution phases of a water-saturated or unsaturated soil is characterized by the \( K_{ow} \) coefficient (i.e., the tendency of a chemical to adsorb). \( K_{ow} \) may be thought of as the ratio of the amount of chemical adsorbed per unit weight of organic carbon in the soil/sediment to the concentration of the chemical in solution at equilibrium, or:

\[
K_{ow} = \frac{mg \text{ adsorbed} / g \text{ organic carbon}}{mg / mL \text{ solution}} \quad (2)
\]

Frequently, the values are fitted to the Freundlich equation and \( K_{ow} \) may range from 1 to 10,000,000 or, on the basis of \( \log K_{ow} \) values, from 0 to 16. FAO (2000) has provided the following criteria to determine the mobility of a chemical in soil according to the adsorption coefficient.

Although no universal indications are available, values of \( \log K_{ow} \) of 5 and above are highly sorptive, meaning that the chemical is likely to be strongly adsorbed to sediments/soils/refuse (Table 3).

(iii) Rate of Biodegradation

The rate of biodegradation is a measure of the speed at which a chemical degrades due to biological processes. The rate is typically measured as a half-life, meaning the time period required for a chemical to reach one-half of the initial concentration. Hence, if a chemical is highly biodegradable (a short half-life), it will likely attenuate within refuse mass in a landfill, as an example, and/or if it is released to the groundwater; attenuate very quickly in the groundwater environment. It is acknowledged that biodegradation rates are highly dependent on whether the ambient environment is aerobic or anaerobic. FAO (2000) criteria provided below is useful to determine the qualitative degradability of the individual substance (Table 4).

(iv) Rate of Vaporization of a Chemical from Water

The rate of vaporization characterizes the degree to which there is movement from the liquid phase to the atmosphere, indicating the potential that a chemical will move, for example, to the biogas phase as opposed to remaining in the aqueous phase and hence ending up in leachate.

A measure of volatilization potential is Henry's Coefficient. Specifically:

If \( H > 2 \times 10^{-5} \text{ atm m}^3/\text{mol} \), volatilization is significant and will, for example, vaporize;

If \( H > 3 \times 10^{-7} \text{ atm m}^3/\text{mol} \), the chemical is highly volatile; and

If \( H < 2 \times 10^{-7} \text{ atm m}^3/\text{mol} \), the chemical is nonvolatile.

FAO (2000) has also provided the following criteria to determine the volatility of the chemical (Table 5).

(v) Solubility

Chemical solubility, the degree to which a compound dissolves in a solvent, is an important factor, where the solubility of a compound in water is given in mg/L at 20 °C. Compounds with high degrees of solubility can be expected to leach into groundwater. FAO (2000) has provided the following criteria to determine the relative solubility of a chemical (Table 6).

(vi) Flashpoint

Knowledge of the flashpoint of a chemical is important in understanding whether a chemical will destruct during combustion processes. A flashpoint of less than 100 °C will, for example, certainly be exceeded during combustion and hence the chemical would be destroyed.

(vii) Disassociation Constant

The pKa or Disassociation Constant is a measure of the strength of an acid or a base. The pKa or ionisation constant is defined as the negative logarithm of the equilibrium coefficient of the neutral and charged forms of a compound. A molecule, or an atom group in a molecule, may lose or gain a proton when the molecule is placed in an aqueous solution. The exact probability that a molecule will be protonated or deprotonated depends on the pKa of the molecule and the pH of the solution. In a focus on aqueous media within the normal environmental pH range of 5 ~ 8, the range of acidities that are of concern is restricted. The acidity range of principal interest corresponds to pKa of 3 to 10. If an organic species has a pKa outside these limits, it expected to be either completely (> 99%) dissociated (pKa of organic acid - < 3) or completely undisassociated (pKa of conjugate acid - > 10) in an aqueous environment.

6. An Example of How the Key Properties Might Be Utilized to Understand the Attenuation Properties of an Emerging Contaminant

As an example how the above information can be used (i.e. the key properties of an emerging contaminant), the potential exists to create an educated characterization of the potential of the emerging contaminant to behave similarly. To demonstrate, consider the following example: an emerging chemical is CHPD having CAS No. of 54079-53-7 and substance name of Propanedinitrile, [[4-[(2-(4-cyclohexylphenoxy)ethyl] ethylamino]-2-methylphenyl)methylene].

CHPD appears to be relatively persistent in the environment, mainly in water, soil and sediments because of its high \( \log K_{ow} \) of 7.88. CHPD is expected to strongly absorb to suspended solids and sediments if released in water, based on its extremely high log Koc value of 6.282. Volatilization from water surfaces is not expected based upon an estimated Henry's Law constant of \( 3.7 \times 10^{-12} \text{ atm-m}^3/\text{mol} \). Given CHPD has a vapour pressure of \( 3 \times 10^{-10} \text{ Pa} \), in combination with Henry's Law Constant, it is reasonable to expect that CHPD is a non-
volatile and therefore is highly unlikely to transfer to the gas phase and may or may not materialize depending upon the pathway.

7. Conclusions

The need for availability of safe water is self-evident, and includes the need to know whether emerging contaminants are present at levels of concern. However, with thousands of new chemicals being introduced every year, and with little information being available about their individual potential to negatively impact humans and the environment, improved procedures need to be identified to improve understanding of fate and transport of emerging contaminants.

The strategy described facilitates the development of a fate and transport exposure pathway assessments to understand which media may contain an emerging contaminant and the degree of attenuation or transformation to another media which may occur. Further, knowledge of the magnitudes of the key coefficients will be useful to determine what type of treatment technology may be effective. To provide these insights, a series of eight key coefficients have been provided which provide guidance whether a specific chemical will attenuate (e.g., biodegrade), guidance on the like-lihood of successful treatment by a particular treatment alternative, or whether attenuation may occur in ambient environmental conditions. Guidance is also available to technical literature which may assist in knowing the carcinogenicity of an emerging contaminant.

While the procedure outlined in the paper is based on properties of key coefficients, it is not without challenges, given the assembly of the many possible behavioral characteristics of an emerging contaminant. However, the approach described herein is based on fundamental characteristics as a mechanism to provide useful insights. Clearly, to manage the task-at-hand to characterize the large number of emerging chemicals, we will have to be as stubborn as Dr. John Snow, as the world cannot afford another PCB or DDT or etc. The Third Water Revolution is already upon us.

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