CHAPTER I

INTRODUCTION

1.1 Pharmaceuticals in the Environment

In the early 1970s, hormones were detected in the environment for the first time (Tabak and Brunch, 1970; Norpoth et al., 1973). In the middle of 1970s, the appearance of pharmaceuticals in the environment was first reported by Garrison et al., 1976. Using GC-MS, they detected clofibric acid in concentrations up to 2 μg/L in the effluent of a municipal sewage treatment plant (STP). Hignite and Azarnoff (1977) found salicylic acid and clofibric acid in the influent and effluent of STP in Kansas City, USA. Over the next decade, other substances (e.g. hydrocarbons, heavy metals, and chlorinated compounds) became of greater interest than pharmaceutical compounds. During the 1990s, public concern over pharmaceuticals in the environment increased greatly. With the advantages in analytical techniques, pharmaceuticals are being detected with increasing frequency in the waters of Europe and USA (Kümmerer, 1997).

At present, pharmaceuticals play an important role for human and animal life. A large amount of pharmaceutical products are produced and used increasingly for human and animal therapy, husbandry, agriculture and aquaculture. Pharmaceutical products are commonly used for illness treatment, prevention of unwanted pregnancy, or facing stress (Zuccato et al., 2000). Besides, they are applied to stimulate a physiological growth in humans, animals, and plants. Several thousand tons of pharmaceutical products were produced and used each year throughout the world.

Unfortunately, some pharmaceuticals can not be metabolized completely during metabolism processes and they are discharged into soil and/or water environments. In addition, they can be run-off from soil after rainfall into the aquatic environment. Furthermore, some pharmaceuticals are not eliminated completely in the municipal sewage treatment plant (STPs) and they are discharged as contaminants into the waters (Heberer, 2002). Pharmaceuticals can enter in the environment by many pathways.

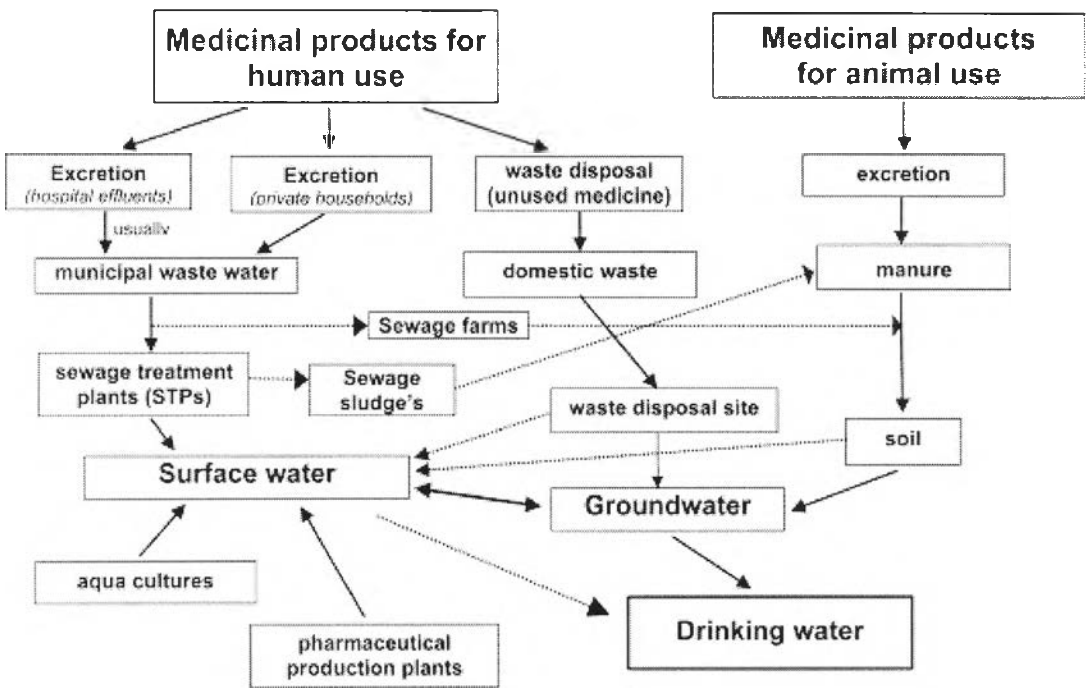

FIGURE 1.1 Possible sources and pathways for the occurrence of pharmaceutical residues in the aquatic environment. (Heberer, 2002)

After the pharmaceuticals enter the environment, some pharmaceuticals are not biodegraded. They can form complexes with metal ions (Chang-Hwa Song et al., 1999) or conjugated with other chemicals and transform into other forms. Some pharmaceuticals can penetrate via organism membrane and accumulate in that organism or absorb into organic material in aquifer soil. These criteria still not to be identified and explored as these interactions are not known.

1.2 Research Aspect

Recently, the appearance of pharmaceuticals in the aquatic environment has become a topic of public interest. Some pharmaceutical compounds cannot be eliminated in wastewater treatment plant and are not degraded in the environment. They have been detected in surface water, groundwater, and drinking water in the ng/L level up to g/L level (Huggett et al., 2003; Heberer, 2002; Kummerer, 2001; Kolpin et al., 2000; Zuccato et al., 2000; Daughton and Ternes, 1999).

Although pharmaceuticals can be detected, information of fate, transport and their nature when they enter the aquatic environment is still limited. Therefore, this study is attempting to evaluate the fate and transport behavior of pharmaceutical compounds. The primary objective of this study is to investigate sorption processes of pharmaceuticals onto pure aquifer materials. Alumina oxide and silica were selected as adsorbent material to mimic the mineral surface in a groundwater aquifer. Porapak P was used to represent the organic adsorbent in the aquatic environment. In this paper, we selected pharmaceuticals for study base on their physicochemical properties, the frequent of detection in the waters, and high level of concentration in the environment. Three different classes of the pharmaceutical compounds that this research focuses are acetaminophen, nalidixic acid, and 17-α-ethynylestradiol.

1.2.1 Objectives

The main purpose of this study is to elucidate the sorption process for acetaminophen, nalidixic acid and 17-α-ethynylestradiol on pure adsorbent materials (silica, alumina, and porapak which are positively charged surface, negatively charged surface and neutral organic material, respectively). Four specific objectives are integrated to accomplish this study:

1. To evaluate sorption kinetics of pharmaceuticals on silica alumina and porapak.
2. To evaluate sorption characteristic of pharmaceuticals on pure adsorbent materials.
3. To investigate the effects of pH on pharmaceutical sorption.
4. To assess the octanol-water partition of pharmaceuticals with varying pH.

1.2.2 Hypotheses

1. Since pharmaceuticals have different molecular structure, the nature of its interaction with adsorbent materials is different.
2. Adsorption of pharmaceutical is a function of pH.
3. The pharmaceuticals sorption characteristic between organic phase and water phase can explain using octanol-water system.

1.2.3 Scopes of the study

Except where specified otherwise, all experiments will be performed with constant ionic concentration, and controlled room temperature. All batch experiments are conducted in laboratory scale.

1. Batch experiments determine adsorption kinetic and sorption isotherm of pharmaceuticals onto pure adsorbent materials.
2. Varying of the pH to determine adsorption characteristic as a function of the pH.
3. Octanol-water partition evaluates the hydrophobicity of pharmaceutical with varying the pH.

1.3 Advantages of Research Studies

Although acetaminophen, nalidixic acid, and 17-α-ethynylestradiol are detected at low concentrations in valuable water systems, work examining their physicochemical properties which affecting the fate and transport of acetaminophen, nalidixic acid, and 17-α-ethynylestradiol has not been reported to date. The interaction of these pharmaceuticals with aquifer materials is unclear. The results of this work will provide new insight and new quantitative tools for evaluating into the fate and transport of acetaminophen, nalidixic acid, and 17-α-ethynylestradiol in groundwater system.