



THIRD EDITION

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# CHEMICAL FATE AND TRANSPORT IN THE ENVIRONMENT

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HAROLD F. HEMOND  
ELIZABETH J. FECHNER



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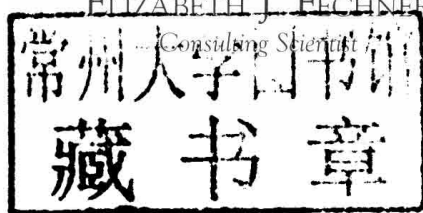
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# Preface

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This textbook is intended for a one-semester course covering the basic principles of chemical behavior in the environment. This third edition has been expanded and extensively revised from the second edition. The common principles that govern both chemical contaminant behavior and the geochemical cycling of naturally occurring chemicals (on which life on Earth depends) are emphasized. The approach is designed to include students who may not be pursuing a degree in environmental science, but whose work will benefit from a basic literacy in environmental transport and fate processes.

Although written as a survey text suitable for graduate students of diverse backgrounds, this book may also be appropriate for use in some undergraduate curricula in environmental engineering. Concepts are developed from the beginning, assuming only prior familiarity with basic freshman chemistry, physics, and math. Certain simplifications are made, and the material is intentionally presented in an intuitive fashion rather than in a rigorously mathematical framework. Nevertheless, the goal is to teach students not only to understand concepts but also to work practical, quantitative problems dealing with chemical fate and transport.

Depending on the nature of the class, the instructor may wish to spend more time with the basics, such as the mass balance concept, chemical equilibria, and simple transport scenarios; more advanced material, such as transient well dynamics, superposition, temperature dependencies, activity coefficients, the thermodynamics of redox reactions, and Monod kinetics, may be omitted. Similarly, by excluding Chapter 4, an instructor can use the text for a course focused only on the water environment. In the case of a more advanced class, the instructor is encouraged to expand on the material; suggested additions include more rigorous derivation of the transport equations, discussions of chemical reaction mechanisms, introduction of quantitative models for atmospheric chemical transformations, use of computer software for more complex chemical equilibrium problems and groundwater transport simulations, and inclusion of case studies. References are provided with each chapter to assist the more advanced student in seeking additional material.

This book was originally based on notes for a class titled Chemicals in the Environment: Fate and Transport, which the first author has taught for over 20 years at the Massachusetts Institute of Technology. Many classes have now used the first and second editions of the textbook; each time, we have benefited from thoughtful feedback from students, teaching assistants, and instructors. We have included many of their suggestions in this

third edition, and hope to hear of the experiences of others who use this text in the coming years. We hope you find the book helpful, even enjoyable, and come away sharing both our enthusiasm for the fascinating environment we inhabit and our desire to treat the environment with appreciation and understanding, not only for its own sake, but to protect the future well-being of humanity worldwide.

*Harry Hemond*

*Liz Fechner*

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## CHAPTER

# 1

## Basic Concepts

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### 1.1 INTRODUCTION

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“By sensible definition any by-product of a chemical operation for which there is no profitable use is a waste. The most convenient, least expensive way of disposing of said waste—up the chimney or down the river—is the best” (Haynes, 1954). This quote describes once-common industrial waste disposal practices, which were based partly on the perception that dispersal of industrial chemical waste into air or water meant that the waste was gone for good. For much of the twentieth century, many industries freely broadcast chemical waste into the environment as a means of disposal. Other human activities, including use of agricultural pesticides, disposal of household waste in landfills, and widespread use of motor vehicles, also contributed enormous loads of anthropogenic chemicals to the environment.

Ultimately, the highly visible, gross pollution effects of emissions from smokestacks and discharge pipes alerted society to the harmful impacts that chemicals could have on the environment and human health. For example, pollutants emitted into the air of industrial cities by factories and automobiles formed smog, resulting in low visibility and increased human mortality. Gross pollution destroyed fisheries and rendered many rivers useless for water supply; in the United States, the Cuyahoga River in Ohio actually caught fire multiple times due to slicks of petroleum-based chemicals. As sources of gross pollution began to be cleaned up, it became evident that severe environmental effects could also be caused by less obvious and less visible chemical releases. For example, the acidification of some lakes by atmospheric deposition of pollutants emitted from power plants, smelters, and automobiles resulted in those lakes losing fish populations. Other lakes burgeoned with unwanted algal growth stimulated by detergent disposal, septic leachate, and urban and agricultural runoff. Drinking water wells were contaminated by chemicals migrating underground from landfills. Human health was put at risk through consumption of fish that had accumulated mercury from the environment via the food chain. Nontoxic, seemingly harmless chlorofluorocarbons (CFCs)

from spray cans and refrigerators eroded Earth's protective ozone shield, thereby threatening the well-being of humans as well as the functioning of ecosystems.

Additional issues regarding potential adverse impacts of chemical releases to the environment continue to be identified. Personal care products (PCPs) and pharmaceuticals now occur widely at detectable levels in natural waters, leading to concern about possible disruption of human endocrine functions, developmental defects in fish and other organisms, and the promotion of antibiotic resistance. Pesticides known as neonicotinoids, used on flowering crops, are being investigated for their potential role in colony collapse of honey bees. New chemicals being developed through nanotechnology are raising concerns about unknown potential toxicities and environmental impacts. And, on the very largest physical scale, a rapidly accumulating body of scientific evidence now confirms that increases in concentrations of carbon dioxide and several trace gases due to anthropogenic activities threaten the stability of Earth's climate.

Despite all these adverse impacts of pollution, it is neither possible nor desirable for modern societies to stop all usage or environmental release of chemicals. Even in prehistoric times, tribes of troglodytes roasting hunks of meat over their fires were releasing complex mixtures of chemicals into the environment. It is imperative, however, that modern societies understand their environment in sufficient detail so that they can make accurate assessments about the environmental behavior and effects of chemicals that they are using. An accurate assessment includes an understanding of both chemical *transport*, referring to processes that move chemicals through the environment, and chemical *fate*, referring to the eventual disposition—either destruction or long-term deposition in the environment—of chemicals. This knowledge is prerequisite if societies are to make intelligent, informed decisions that will protect both human health and the environment, while allowing human beings to enjoy the benefits of modern technology.

Traditionally, the discipline of environmental engineering has addressed anthropogenic pollutant issues, such as pollution prevention and site cleanup, whereas natural chemical cycles, such as nutrient cycling, have been the domain of ecology and environmental chemistry. However, there are numerous natural chemical cycles upon which humans and all other life forms depend for their existence, and many human activities, including direct pollutant emissions, affect these cycles. Therefore, it is important to understand the fate and transport processes associated with natural chemical cycles as well as with anthropogenic pollutants. In this book, some key natural chemical cycles are described along with prominent pollution problems; the principles governing fate and transport can be applied equally. Note that anthropogenic pollutants include naturally occurring chemicals whose loadings to the environment have been greatly

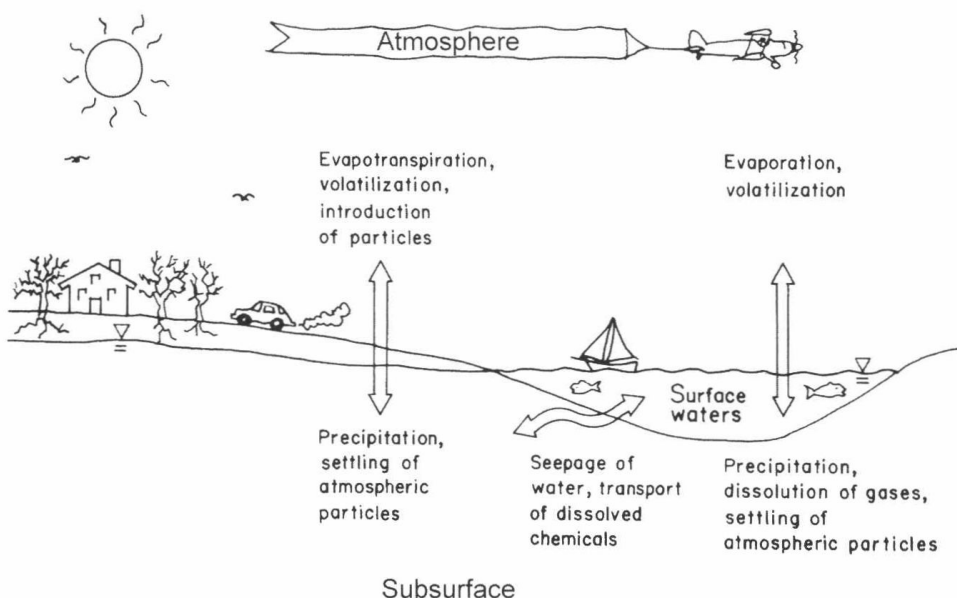
increased by human activities, as well as *xenobiotic* (foreign to life) chemicals that do not occur naturally in the environment.

The principles that govern the fate and transport of chemicals are presented for three major environmental media: surface waters, soil and groundwater (the subsurface), and the atmosphere. These several media are treated in one book for three related reasons. First, this is primarily an introductory textbook, and a broad scope is most appropriate for the student who has not specialized in a particular environmental medium. Second, chemicals released into the environment do not respect the boundaries between air, water, and soil any more than they respect political boundaries. Because exchanges among these media are common, modeling a chemical in any single medium is unlikely to be adequate for obtaining a full description of the chemical's fate. Third, a great deal of insight can be gained by comparing and contrasting chemical behavior in surface waters, soil and groundwater, and the atmosphere. For example, although the fact is not immediately apparent in much of the scientific literature, the mathematics describing physical transport in each medium are almost identical; the transport equation that models the mixing of industrial effluent into a river is also useful for describing the movement of chemicals from a leaking underground tank, or the mixing of smoke-stack emissions into the atmosphere. Contrasts are also instructive; for example, the dominant fate process for a chemical in the atmosphere may be photodegradation, whereas in soil and groundwater, biodegradation of the chemical is more likely to dominate.

Knowledge of the principles underlying the fate and transport of chemicals in the environment allows problems ranging from local to global scales to be defined and analyzed. This first chapter presents fundamental concepts that apply universally to any environmental medium. The subsequent three chapters focus on surface waters, the subsurface, and the atmosphere, respectively; see Fig. 1.1 for a diagram of some of the interrelationships among these media. In each chapter, each medium is discussed in terms of its basic physical, chemical, and biological attributes, and then the fate and transport of chemicals within the medium are considered.

## 1.2 CHEMICAL CONCENTRATION

Perhaps the single most important parameter in environmental fate and transport studies is chemical *concentration* ( $C$ ). The concentration of a chemical in the environment is a measure of the amount of that chemical in a specific volume or mass of air, water, soil, or other material. A chemical's concentration in an environmental medium not only is a key quantity



**FIGURE 1.1** Three major environmental media: surface waters, the subsurface, and the atmosphere. Although each medium has its own distinct characteristics, there are also many similarities among them. Few chemicals are restricted in their movement to only one medium; thus chemical exchanges among the media must be considered. Several very generalized exchange processes between media are shown.

in fate and transport equations but also partly determines the magnitude of its biological effect on ecosystems and human health.

Most laboratory analysis methods (see Section 1.8.1) measure concentration. The choice of units for concentration depends on the medium, on the process that is being measured or described, and sometimes on custom and tradition. In water, a common expression of concentration is mass of chemical per unit volume of water, which has dimensions of  $[M/L^3]$ . The letters M, L, and T in square brackets refer to the fundamental dimensions of mass, length, and time (see Appendix). Many naturally occurring chemicals in water are present at levels of a few milligrams per liter (mg/liter). For clarity in this book, specific units, such as (cm/hr) or (g/m<sup>3</sup>), either are free-standing or are indicated in parentheses, not in square brackets. Note that the word "liter" is always spelled out in this text, to avoid confusion with the abbreviation [L] for the fundamental dimension length.

Another common unit of concentration in water is *molarity*. A *mole* of a chemical substance is composed of  $6.02 \times 10^{23}$  atoms or molecules of that substance. (The number of atoms in a mole is sometimes referred to as *Avogadro's number*.) Molarity refers to the number of moles per liter of aqueous solution (mol/liter); in this book, molarity is denoted by *M*, with neither parentheses nor square brackets around it.

A related unit, *normality* ( $N$ ), refers to the number of *equivalents* of a chemical per liter of water. An equivalent is the amount of a chemical that either possesses, or is capable of transferring in a given reaction, 1 mol of electronic charge. If a chemical has two electronic charge units per molecule, 1 mol of the chemical constitutes two equivalents (e.g., a mole of sulfate ( $\text{SO}_4^{2-}$ ) is equal to two equivalents, and a one molar (1  $M$ ) solution of sodium sulfate ( $\text{Na}_2\text{SO}_4$ ) is two normal (2  $N$ )).

In soil, a chemical's concentration may be measured in units such as milligrams per cubic centimeter ( $\text{mg}/\text{cm}^3$ ). Expressing concentration as mass per unit volume for soil, however, carries the possibility of ambiguity; soils undergo volume changes if they are compacted or expanded. Accordingly, it is often more useful to express soil concentration as chemical mass per unit mass of soil (e.g.,  $\text{mg}/\text{kg}$ ), because the mass of soil does not vary with changes in the degree of compaction of the soil.

For air, which is highly compressible, expression of chemical concentration as mass per volume can also be ambiguous. The volume of a given mass of air changes significantly with changes in pressure and temperature, and thus the chemical concentration, when expressed as mass per volume, also changes. For example, consider a *vapor*, a chemical in gaseous form, which may coexist with the chemical's liquid or solid phase at ordinary temperatures and pressures. If initially there is 1  $\mu\text{g}$  of vapor per cubic meter of air and the barometric pressure decreases by 5%, the concentration of vapor as expressed in units of mass per volume also decreases by 5%, because the air expands while the mass of gaseous chemical remains constant. Expressing the vapor concentration as mass of gaseous chemical per mass of air removes ambiguity caused by temperature and pressure effects. It is equally valid, and more common, to express a gaseous chemical concentration in air as a volume ratio, e.g., volume of vapor per volume of air. In this case, a pressure decrease of 5% results in equal expansion of the air and the gaseous chemical, and thus there is no change in their volume ratio.

Ambiguity can also arise when a laboratory reports the concentration of a chemical that can exist in more than one form (*species*) without reference to a particular species. For example, if the concentration of nitrogen in a water sample is reported simply as 5  $\text{mg}/\text{liter}$ , it is unclear whether 1 liter contains 5  $\text{mg}$  of nitrogen atoms ( $N$ ) or 5  $\text{mg}$  of one of the nitrogen species present, such as nitrate ( $\text{NO}_3^-$ ), nitrite ( $\text{NO}_2^-$ ), ammonia ( $\text{NH}_3$ ), or ammonium ( $\text{NH}_4^+$ ). In such a situation, clarification must be obtained from the analytical laboratory regarding the actual chemical species measured. The species associated with a concentration must be known to convert from mass to moles; the number of moles equals the chemical's mass in grams divided by the chemical's *molecular weight*, i.e., the number of grams per mole of the chemical.



Numerous other options exist for specifying concentration; common ones are parts per thousand (ppt or ‰), parts per million (ppm), or parts per billion (ppb). For soil or air, ppm on a mass basis is numerically equal to milligrams (mg) of chemical per kilogram (kg) of soil or air, respectively. Parts per million on a volume basis, denoted ppm(v), is typically used for air; 1 ppm(v) of helium in air would correspond to 1 milliliter (ml) of helium in 1000 liter ( $1 \text{ m}^3$ ) of the air-helium mixture. For water, the density of which is approximately  $1 \text{ g/cm}^3$ , parts per million corresponds to milligrams of chemical per liter of water (mg/liter) in dilute solutions. Be cautious of the opportunities for confusion when units of ppt, ppm, or ppb are used! The use of actual units for concentration removes all ambiguity. Clarity in the expression of chemical concentration is critical to the implementation of a central concept of environmental fate and transport: being able to account for all the mass of a chemical in a given environmental setting.

## 1.3 MASS BALANCE AND UNITS

### 1.3.1 Mass Balance and the Control Volume

Three possible outcomes exist for a chemical present at a specific location in the environment at a particular time: the chemical can remain in that location, the chemical can be carried elsewhere by a transport process, or the chemical can be eliminated or produced through *transformation* into or from another chemical. This very simple observation is known as *mass balance* or *mass conservation*. Mass balance is a concept around which an analysis of the fate and transport of any environmental chemical can be organized; mass balance also serves as a check on the completeness of knowledge of a chemical's behavior. If, at a later time in an analysis, the original mass of a chemical cannot be fully accounted for, then there is an incomplete understanding of the transport and transformation processes affecting that chemical.

Implicit in the application of the mass balance concept is the need to choose a *control volume*. A control volume is any defined volume within which all the chemical initially present (stored) and all processes that internally produce or eliminate the chemical can be accounted for, and across whose boundaries all chemical transport can, in principle, be quantified. The *mass balance expression* (also sometimes called the *material balance expression*) for any chemical in a control volume during any given time interval can be written as

$$\begin{aligned} \text{Change in storage of mass} = & \text{mass transported in} - \text{mass transported out} \\ & + \text{mass produced by internal sources} \\ & - \text{mass eliminated by internal sinks.} \end{aligned}$$

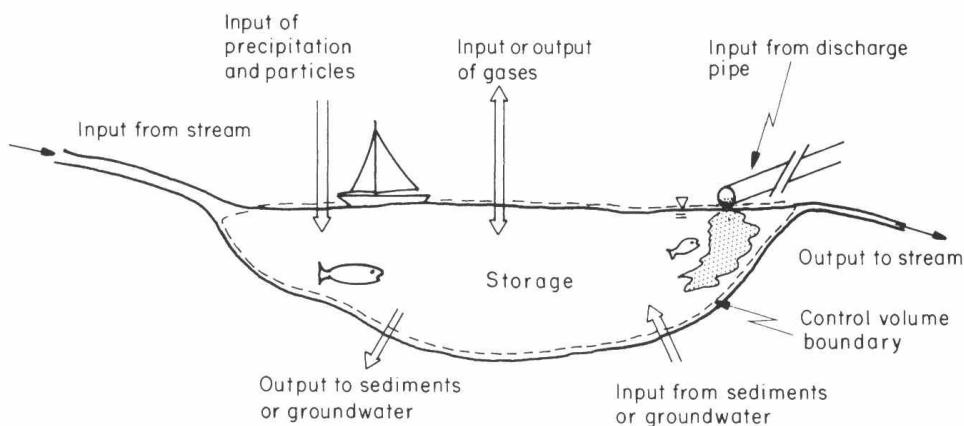
(1.1a)

The mass balance expression in a control volume can also be written in terms of rates, that is, mass per time  $[M/T]$ :

$$\begin{aligned} \text{Rate of change in storage of mass} &= \text{mass transport rate in} \\ &\quad - \text{mass transport rate out} \\ &\quad + \text{mass production rate by internal sources} \\ &\quad - \text{mass elimination rate by internal sinks.} \end{aligned} \quad (1.1b)$$

Control volumes are chosen to be convenient and useful. While the choice of a good control volume is somewhat of an art and depends on both the chemicals and the environmental locations of interest, control volume boundaries are almost always chosen to simplify the problem of determining chemical transport into and out of the control volume.

As an example of an environmental pollution problem requiring the choice of a control volume, consider a lake that is receiving industrial effluent from a discharge pipe. To establish a useful control volume within which the fate and transport of the effluent chemicals can be described, the entire lake, as shown in Fig. 1.2, could be chosen. The upper boundary of this control volume is the lake surface; transport across this boundary is described by principles that govern chemical transport between water and air. (Transport of a chemical from water to air is one example of *volatilization*.) For chemicals that preferentially remain dissolved in water and have little tendency to escape to the atmosphere, this transport rate is negligible, but for chemicals for which air-water transfer is significant, it is necessary to have information on both chemical properties and on the physics of transport across the lake surface. The lower boundary of this control volume might be chosen to be slightly above the bottom sediments



**FIGURE 1.2** An example of a control volume that is useful for estimating chemical mass balance in a lake. Chemicals may enter the control volume via the inflowing stream, the industrial discharge pipe, the sediment–water interface, and precipitation. Chemicals may leave the control volume across the air–water interface and the sediment–water interface as well as via the outflowing stream.