J. Hrubec
Editor

Quality and Treatment of Drinking Water

The Handbook of Environmental Chemistry

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Edited by O. Hutzinger

Water Pollution

Drinking Water and Drinking Water Treatment

Volume Editor: J. Hrubec

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Preface

Environmental Chemistry is a relatively young science. Interest in this subject, however, is growing very rapidly and, although no agreement has been reached as yet about the exact content and limits of this interdisciplinary subject, there appears to be increasing interest in seeing environmental topics which are based on chemistry embodied in this subject. One of the first objectives of Environmental Chemistry must be the study of the environment and of natural chemical processes which occur in the environment. A major purpose of this series on Environmental Chemistry, therefore, is to present a reasonably uniform view of various aspects of the chemistry of the environment and chemical reactions occurring in the environment.

The industrial activities of man have given a new dimension to Environmental Chemistry. We have now synthesized and described over five million chemical compounds and chemical industry produces about one hundred and fifty million tons of synthetic chemicals annually. We ship billions of tons of oil per year and through mining operations and other geophysical modifications, large quantities of inorganic and organic materials are released from their natural deposits. Cities and metropolitan areas of up to 15 million inhabitants produce large quantities of waste in relatively small and confined areas. Much of the chemical products and waste products of modern society are released into the environment either during production, storage, transport, use or ultimate disposal. These released materials participate in natural cycles and reactions and frequently lead to interference and disturbance of natural systems.

Environmental Chemistry is concerned with *reactions in the environment*. It is about distribution and equilibria between environmental compartments. It is about reactions, pathways thermodynamics and kinetics. An important purpose of this Handbook is to aid understanding of the basic distribution and chemical reaction processes which occur in the environment.

Laws regulating toxic substances in various countries are designed to assess and control risk of chemicals to man and his environment. Science can contribute in two areas to this assessment: firstly in the area of toxicology and secondly in the area of chemical exposure. The available concentration ("environmental exposure concentration") depends on the fate of chemical compounds in the environment and thus their distribution and reaction behaviour in the environment. One very important contribution of Environmental Chemistry to the above mentioned toxic substances laws is to develop laboratory test methods, or mathematical correlations and models that predict the environmental fate of new chemical compounds. The third purpose of this Handbook is to help in the basic understanding and development of such test methods and models.

The last explicit purpose of the handbook is to present, in a concise form, the most important properties relating to environmental chemistry and hazard assessment for the most important series of chemical compounds.

At the moment three volumes of the Handbook are planned. Volume 1 deals with the natural environment and the biogeochemical cycles therein, including some background information such as energetics and ecology, Volume 2 is concerned with reactions and processes in the environment and deals with physical factors such as transport and adsorption, and chemical, photochemical and biochemical reactions in the environment, as well as some aspects of pharmacokinetics and metabolism within organisms. Volume 3 deals with anthropogenic compounds, their chemical backgrounds, production methods and information about their use, their environmental behaviour, analytical methodology and some important aspects of their toxic effects. The material for volumes 1, 2, and 3 was more than could easily be fitted into a single volume, and for this reason, as well as for the purpose of rapid publication of available manuscripts, all three volumes are published as a volume series (e.g. Vol. 1; A, B, C). Publisher and editor hope to keep the material of the volumes 1 to 3 up to date and to extend coverage in the subject areas by publishing further parts in the future. Readers are encouraged to offer suggestions and advice as to future editions of "The Handbook of Experimental Chemistry".

Most chapters in the Handbook are written to a fairly advanced level and should be of interest to the graduate student and practising scientist. I also hope that the subject matter treated will be of interest to people outside chemistry and to scientists in industry as well as government and regulatory bodies. It would be very satisfying for me to see the books used as a basis for developing graduate courses on Environmental Chemistry.

Due to the breadth of the subject matter, it was not easy to edit this Handbook. Specialists had to be found in quite different areas of science who were willing to contribute a chapter within the prescribed schedule. It is with great satisfaction that I thank all authors for their understanding and for devoting their time to this effort. Special thanks are due to the Springer publishing house and finally I would like to thank my family, students and colleagues for being so patient with me during several critical phases of preparation for the Handbook, and also to some colleagues and the secretaries for their technical help.

I consider it a privilege to see my chosen subject grow. My interest in Environmental Chemistry dates back to my early college days in Vienna. I received significant impulses during my postdoctoral period at the University of California and my interest slowly developed during my time with the National Research Council of Canada, before I was able to devote my full time to Environmental Chemistry in Amsterdam. I hope this Handbook will help deepen the interest of other scientists in this subject

This preface was written in 1980. Since then publisher and editor have agreed to expand the Handbook by two new open-ended volume series: Air Pollution and Water Pollution. These broad topics could not be fitted easily into the headings of the first three volumes.

All five volume series will be integrated through the choice of topics covered and by a system of cross referencing.

The outline of the Handbook is thus as follows:

1. The Natural Environment and the Biogeochemical Cycles,
2. Reactions and Processes,

3. Anthropogenic Compounds,
4. Air Pollution,
5. Water Pollution.

Fifteen years have passed since the appearance of the first volumes of the Handbook and four years since the last preface. Our original concept of collecting solid scientific information in Environmental Chemistry has been well received, and with the help of many authors and volume-editors we have published a total of 24 books.

Although recent emphasis on chemical contaminants and industrial processes has broadened to include toxicological evaluation, risk assessment, life cycle analysis and similar approaches there is still a need for presentation of chemical and related facts pertaining to the environment. The publisher and editor therefore decided to continue our five volume series.

Bayreuth, January 1995

Otto Hutzinger

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Introduction

In the last decades, contamination of drinking water and growing public concern about the health risks of contaminants have received much publicity and initiated many research efforts, as well as political and legal activities.

The majority of the recent problems related to drinking water contamination, associated with pollution of surface and ground water resources and with the formation of reaction by-products resulting from the use of disinfectants and oxidants in drinking water treatment, is closely connected with the rapid advances in analytical techniques. The modern analytical methods have resulted in the identification of a large number of chemical compounds and microbial pollutants since the early seventies. Continuing discoveries of new drinking water pollutants and related health hazards have had a shocking effect on the public. For the professional community they have created a multitude of unknown factors and uncertainties concerning toxicological, technological and regulatory aspects.

One of the major issues related to drinking water contamination is the assessment of the health hazards and associated risk comparisons, priority settings and risk management. The health hazard assessment plays an important role in the evaluation of the overall relevance of the problem and is one of the principal factors in the formulation of research needs. A specific feature of health hazards related to drinking water contamination constitutes a dilemma of "competing risk", leading to reduction of a "target risk" and simultaneously creating other kinds of risks. A well known example is the use of chemical disinfectants for elimination of microbial risk, resulting in an increase of health risks from the formation of reaction by-products and vice versa. As a result reduction of risk from formation of by-products by restrictive measures in the application of chemicals can result in an increase of microbial risk.

Health risk assessment has a decisive influence on the setting of national and international quality standards and directives. Due to the current limited state of scientific knowledge and the complexity of political and social reality the quality standards have only a temporary character and therefore constitute an unstable, but nevertheless the only available rational basis for the formulation of technological and technical goals and objectives.

As far as treatment of drinking water is concerned, since 1974, when the formation of trihalomethanes by chlorination was discovered, chlorination by-products are the major research topic. A large number of studies on identification of the reaction by-products of chlorination and on their toxicological effects has provided convincing reasons for avoiding the use of chlorine in drinking water treatment and for the use of alternative disinfection methods.

However, much less information is available on the consequences of the application of alternatives for chlorine, such as ozone and chlorine dioxide. Still, insufficient evidence exists that the reaction by-products of alternative disinfectants and oxidants are less hazardous than those of chlorine. An important warning, which can be learned from the research on alternative disinfectants for drinking water treatment, is the fact, that the application of any "transformation" process in drinking water treatment introduces a high risk of formation of by-products, which are currently largely unidentifiable and have unknown health effects. Clearly a preference should be given to "real removal" processes, such as aeration, adsorption on activated carbon and membrane separation. Considerable progress has been made recently in the understanding and in the practical application of these processes.

The most serious threat for drinking water quality indeed is posed by the pollution of drinking water resources. As far as surface water is concerned it is caused by anthropogenic compounds, by pathogenic microorganisms and by pollutants related to eutrophication, such as odor and taste compounds and algae toxins.

Ground water, traditionally considered as the most safe drinking water source, has been threatened more and more by the waste dumping, by nitrate and pesticides, resulting from agricultural activities and from air pollution.

Finally still more attention is being given to the quality deterioration of drinking water during transportation and storage as a result of material corrosion and biological activity promoted by the presence of biodegradable compounds.

This volume does not attempt to be an exhaustive review of such a vast and complex subject as drinking water quality, but it is meant to give an overview of the developments in key areas related to chemical contamination, with special attention to organic micropollutants.

The two parts of the volume are organized as follows:

The first part principally addresses:

- The latest developments in quality regulation.
- The role of biological processes in degradation of organic micropollutants and in control of biological instability of drinking water.
- Significance of biological stability of drinking water.
- Control of organic micropollutants by adsorption on activated carbon.
- Origin and removal of tastes and odors.

The second part of the volume will focus mainly on identification of organic micropollutants, approaches to the evaluation of health hazards from chemical and microbiological pollution, the issue of algae toxins, the threat posed to groundwater quality by contamination from agricultural activities and quality changes due to application of ozone and chlorine dioxide.

From the important drinking water quality issues the volume does not address microbiological pollution, because of the scope of the Handbook. From the chemical issues, the principal topic of the reaction by-products of chlorination is not addressed, mainly because it is covered in great detail in a number of other publications. One of the basic aspects of the chlorination problem -health risks of chlorinated drinking water- has been already reviewed in the Handbook elsewhere (see Craun GF Vol. 5, Part A, p 1).

Statutory and Regulatory Basis for Control of Drinking Water Quality

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List of Symbols and Abbreviations

ADI Acceptable Daily Intake (mostly used for food additives)
BAT Best Available Technology
D-DBP Disinfectant-Disinfection Byproduct
EC European Community
EUREAU Union of National Associations of Water Suppliers from countries within the European Community and the Economic Free Trade Association
GL Guide Level
ICR Information Collection Rule
MAC Maximum Admissible Concentration
MCL Maximum Contaminant Level
MCLG Maximum Contaminant Level Goal
QSAR Quantitative Structure Activity Relationship

SMCL Secondary Maximum Contaminant Level
SWTR Surface Water Treatment Rule
TDI Tolerable Daily Intake (mostly used for contaminants which cannot be avoided)
USEPA United States Environmental Protection Agency
WHO World Health Organization

Abstract

Drinking water standards and regulations have recently been developed and revised both at national and international level. The three main topics discussed in this article are the revision of the EC Drinking Water Directive (80/778/EEC), the revision of the 1984 WHO Guidelines for Drinking Water Quality and the standard setting procedure of the USEPA according to the 1986 amendments of the Safe Drinking Water Act. For future regulation of drinking water quality, the importance is stressed of:

a) integrated standards for human exposure to chemicals;
b) informing the public of health risks due to exposure to hazards via drinking water, food, air and skin.

The importance of effective monitoring and appropriate treatment techniques are also discussed.

Introduction

The first drinking water standards were issued probably more than 4000 years ago. Baker [1] quotes from a Sanskrit source: "... it is directed to heat foul water by boiling and exposing to sunlight and by dipping seven times into a piece of hot copper, then to filter and cool in an earthen vessel." During the development of water treatment processes as we know them today, in the last 150 years drinking water quality standards have evolved considerably.

Until 1980 the individual European countries had different regulations with standards covering about 24 parameters which were partly based on European [2] and International [3] standards for drinking water. In 1980, the EC Drinking Water Directive was issued with standards covering 62 parameters. This directive has been implemented in the national legislation of the Member States in subsequent years.

In the USA a standard covering coliforms was introduced in 1914 for protection of the traveling public, followed by standards covering other physical and chemical constituents in 1925. In 1943, 1946 and 1962 parameters were added and in 1974 the Public Law (Safe Drinking Water Act) passed Congress which allowed EPA to promulgate drinking water standards, which led to a list of 83 compounds in 1986 to be regulated or reevaluated.

WHO published its Guidelines for drinking water quality in 1984. For 40 physicochemical parameters and for microbiological parameters and radioactivity, guidelines were given [4].

In Russia, standards for drinking water have been set for 29 parameters but proposals for adding 40 new parameters are being discussed [5].

So looking back only a few decades it is clear that the number of standards and regulations has grown fast.

The ongoing progress in analytical chemistry, the growing awareness of the threats to our environment, the increase in industrial activities and the availability of toxicological data have recently resulted in (starting) a new revision of drinking water standards and regulations both on a national and international level. Three main topics in this area are:

1. the revision of the EC Drinking Water Directive (80/778/EEC);
2. the revision of the 1984 WHO Guidelines for Drinking Water Quality;
3. the ongoing standard setting procedure of the USEPA, according to the 1986 Amendments of the Safe Drinking Water Act.

These topics will also be important for the many new countries in Europe which will have to develop their own national drinking water standards in the near future.

Looking to the future, many questions may arise when setting and implementing standards, for instance:

- will standards and regulations provide a good quality drinking water under all circumstances?
- will we end up with regulations of hundreds of chemicals in drinking water?
- should we stimulate the public's knowledge of the basic ideas of standard setting and risk perception?

It will take some time before all the questions are answered and implemented in day to day practice. Nonetheless it seems worthwhile to stimulate discussion on these matters.

Revision of the EC Drinking Water Directive (80/778/EEC)

The present Directive was developed in the early seventies and was adapted in 1980. In subsequent years it has been implemented in the legislation of the European Member States. In the Drinking Water Directive (80/778/EEC) MACs (maximum admissible concentrations) have been set for 41 parameters. For 12 parameters, only guide levels (GL) have been set and for 16 parameters both a MAC and a GL (see Annex I). The values for the parameters to be fixed by the Member States should be less than or equal to the MAC value and the Member States should take the levels appearing in the "Guide level column" as a basis.

A summary of the way in which the directive is implemented in the different Member States is given by Premazzi et al. [6]. Apart from emergency situations it is not possible to derogate from the MAC values for toxic or microbiological parameters. For the other parameters derogations from the Directive are only possible when:

- the nature and structure of the ground in the area from which the supply is taken into account;
- exceptional meteorological conditions arise.

There is no doubt that the Drinking Water Directive has made a valuable contribution to the recognition of the importance of drinking water quality and has been a trigger for improvements in water treatment. Recently, however, many Member States and other important parties have asked for a revision of the directive, based on the following:

1. in the past ten years progress has been made in technical and scientific understanding of water quality;
2. a number of points in the directive are not clear or should be modified;
3. several analytical methods given in the directive are not unambiguously defined;
4. the basis for the standards in the directive is not laid down. There are no health criteria documents available. This leads to confusion and a lot of questions when large investments are involved in order to comply with the standards (for example nitrate and pesticides).

In anticipation of the above, the Commission has opened a discussion on the general problems of the Member States associated with the implementation of the directive and for the exchange of ideas for possible modifications. Most delegations were in favour of updating the Directive, although there was still some controversy.

In 1991, EUREAU (The Union of National Associations of Watersuppliers from countries within the European Community and the Economic Free Trade Association) developed proposals for improvements and modifications of the directive [7]. These proposals have recently been updated [8]. A summary of the EUREAU proposals is given in Table 1.

At the third European conference of EUREAU (March, 1993), the Commission (via Garvey, Deputy Director General of DG XI), announced that a revision process of the directive would be initiated. In September 1993 a conference has been organized by the EC to make an inventory of the wishes of both the Member States and other interested parties (EUREAU, environmentalists, industrialists). The Commission has already made clear that [9]:

1. the revision process should include the results of the most recent scientific investigations;
2. the perception of the consumer regarding drinking water must be considered more extensively;
3. not only health aspects but also taste and other aesthetic/organoleptic aspects are important;
4. the consumer must be able to trust or regain trust in drinking water from the tap with regards to health and taste;

Table 1. Part of the EUREAU proposals for modification of the EC Drinking Water Directive

- The classification of the parameters in the present directive should be changed in such a way that it is easier to lay down criteria of exceedances and analytical obligations. Considering that the main objective is the protection of the health and comfort of the consumer EUREAU suggests dividing the parameters into health related (microbiological and chemical), aesthetic/organoleptic and operational.
- The revision should reassess the basis and use of Maximum Admissible Concentration. Limits should be based on the most recent scientific knowledge taking into account the work of international bodies such as WHO. In setting limits for health-related parameters account should be taken of the fact that WHO Guidelines are generally based on lifetime consumption and are not therefore equivalent to MACs.
- Operational parameters have no direct effect on the health of the consumers. They are operational indicators to achieve optimal drinking water conditions and are often highly valuable depending on local circumstances. Examples of these parameters are: temperature, pH, conductivity, chlorides, TOC and total bacteria counts in supplied water. Mandatory standards for these parameters should not be set by the EC, but on a national or regional level.
- The revision should evaluate all parameters in the current Directive. A number of parameters should be deleted from the directive because they are either not relevant to the drinking water quality (silica, potassium, Kjeldahl nitrogen) or covered by others (total hardness, dry residues, suspended solids).
- The addition of parameters should be subject to very careful consideration in view of the practical and financial implications.
- Guide levels should be removed, since they have no scientific basis and have often led to confusion on the part of the consumer.
- Procedures need to be developed to deal with exceedance of the limits. These procedures should take into account the nature of the parameters and the circumstances of the exceedance. For nonhealth-related parameters (for example colour) compliance rules and criteria for allowing for exceedances due to local natural conditions should be developed.
- Legislation for protecting water sources should be reviewed to provide a raw water that allows its use for drinking water without enhanced treatment. This will result in compliance with the Drinking Water Directive, thus lessening the reliance on water treatment. This will lead to more cost-effective consumer protection.

5. the EC will not tolerate a weakening of the directive;
6. an equilibrium must be found between a flexible application of the directive and the level of protection of the consumer;
7. financial implications should be taken into account when setting new standards.

Although the leeway for a revision of the directive is small in view of the above, the EC will have to ensure a sound basis for the standards due to the legal and financial implications involved.

WHO Guidelines for Drinking Water Quality

In 1984, the World Health Organisation (WHO) published its Guidelines for Drinking Water Quality [4]. These Guidelines are intended for use as a basis for the development of standards which, if properly implemented, will ensure the safety of drinking water. It must be stressed that the WHO guidelines have to be interpreted in the correct way; the guideline values must be considered in the context of prevailing environmental, social, economic and cultural conditions.

Some important aspects of the nature of the WHO guideline values are:

1. a guideline value represents the level (a concentration or a number) of a constituent, that ensures an aesthetically pleasing water and does not result in any significant risk to the health of the consumer over a lifetime's consumption;
2. short-term deviations above the guideline values do not necessarily mean that the water is unsuitable for consumption. The amount by which and the period for which any guideline value can be exceeded without affecting public health depends on the specific substance involved;
3. when setting or developing national standards on the basis of the guidelines, it is necessary to take account of geographical, socio-economic, dietary and other conditions affecting potential exposure. This may lead to national standards which differ appreciably from the guideline values.

The information used for the 1984 Guidelines dates from 1980 or earlier. In 1988 it was decided that the Guidelines should be revised. Over a period of 4 years, 14 meetings were organized by WHO at which 127 chemical compounds as well as the microbiological parameters and radio-activity were evaluated.

The revision of the Guidelines was completed in September 1992 and Volume 1 of the revised Guidelines was published in November 1993 [10].

A few principles of the derivation of the guideline values are important:

1. the guideline values are based on toxicity data. For the majority of the substances for which guideline values are proposed the toxic effect in humans is predicted from studies with laboratory animals. In extrapolating such animal data to humans, safety factors or mathematical methods are used, depending on the toxicity of the compounds involved;
2. each country may choose its own risk level when mathematical models are used to derive a guideline value for a genotoxic carcinogen. As an example WHO has chosen a risk level of 10^{-5} at lifetime consumption;
3. WHO stresses that the guideline values of disinfectants and disinfectant byproducts may not influence the microbiological quality of the water. The microbiological quality of the water has a much higher priority;
4. some guideline values are still provisional. The term provisional guideline value is used for:
   - compounds for which there is some evidence of a potential hazard but where the available health effects information is limited; and/or where an uncertainty factor larger than 1000 is used in the derivation of the tolerable daily intake;
   - those substances for which the calculated guideline value based on toxicological information would be (a) below the practical quantification level or (b) below the level that can be achieved through practical

treatment methods or where disinfection is likely to result in the guideline value being exceeded;

5. in contrast with the guideline values of 1984, WHO has not given guideline values with regard to the aesthetic/organoleptic quality of the water. Numerical guidelines for acceptability aspects were found to be undesirable because of the danger of misinterpretation. When, for example, water sources are scarce, highly coloured drinking water may be refused although, from a health point of view, the drinking water can be consumed. In the revised Guidelines WHO only provides a table with values for aesthetic/organoleptic parameters, which may give rise to complaints from consumers. These values are given in Table 2 together with the nonenforceable USA federal guideline values, referred to as Secondary Maximum Contaminant Levels (SMCLs). The revised guideline values are given in Annex I.

In comparison with the WHO 1984 guidelines, 86 new parameters have been introduced. From these 86 guidelines values, 14 are still provisional.

Table 2. Substances and parameters in drinking water that may give rise to complaints from consumers [10] and SMCLs of the USEPA.

	WHO		USEPA
	Levels likely to give rise to consumer complaints[1]	Reasons for consumer complaints[2]	SMCL
A. Inorganics			
aluminium	0.2 mg/l	depositions, discoloration	0.05–0.2 mg/l
ammonia	1.5 mg/l	odour and taste	–
chloride	250 mg/l	taste, corrosion	250 mg/l
colour	15 TCU	appearance	15 TCU
copper	1 mg/l	staining of laundry and sanitary ware (health-based provisional GV 2 mg/l)	1 mg/l
hardness	–	high hardness: scale deposition, scum formation low hardness: possible corrosion	–
hydrogen sulfide	0.05 mg/l	odour and taste	–
iron	0.3 mg/l	staining of laundry and sanitary ware	0.3 mg/l
manganese	0.10 mg/l	staining of laundry and sanitary ware (health-based provisional GV 0.5 mg/l)	0.05 mg/l
dissolved oxygen	–	indirect effects	–
pH	–	low pH: corrosion high pH: taste, soapy feel preferably < 8.0 for effective disinfection with chlorine	6.5–8.5