**WHO/SDE/WSH/03.04/73**
**English only**

# Aldrin and Dieldrin in Drinking-water

Background document for development of
WHO *Guidelines for Drinking-water Quality*

## Preface

One of the primary goals of WHO and its member states is that "all people, whatever their stage of development and their social and economic conditions, have the right to have access to an adequate supply of safe drinking water." A major WHO function to achieve such goals is the responsibility "to propose ... regulations, and to make recommendations with respect to international health matters ...."

The first WHO document dealing specifically with public drinking-water quality was published in 1958 as *International Standards for Drinking-water*. It was subsequently revised in 1963 and in 1971 under the same title. In 1984–1985, the first edition of the WHO *Guidelines for Drinking-water Quality* (GDWQ) was published in three volumes: Volume 1, Recommendations; Volume 2, Health criteria and other supporting information; and Volume 3, Surveillance and control of community supplies. Second editions of these volumes were published in 1993, 1996 and 1997, respectively. Addenda to Volumes 1 and 2 of the second edition were published in 1998, addressing selected chemicals. An addendum on microbiological aspects reviewing selected microorganisms was published in 2002.

The GDWQ are subject to a rolling revision process. Through this process, microbial, chemical and radiological aspects of drinking-water are subject to periodic review, and documentation related to aspects of protection and control of public drinking-water quality is accordingly prepared/updated.

Since the first edition of the GDWQ, WHO has published information on health criteria and other supporting information to the GDWQ, describing the approaches used in deriving guideline values and presenting critical reviews and evaluations of the effects on human health of the substances or contaminants examined in drinking-water.

For each chemical contaminant or substance considered, a lead institution prepared a health criteria document evaluating the risks for human health from exposure to the particular chemical in drinking-water. Institutions from Canada, Denmark, Finland, France, Germany, Italy, Japan, Netherlands, Norway, Poland, Sweden, United Kingdom and United States of America prepared the requested health criteria documents.

Under the responsibility of the coordinators for a group of chemicals considered in the guidelines, the draft health criteria documents were submitted to a number of scientific institutions and selected experts for peer review. Comments were taken into consideration by the coordinators and authors before the documents were submitted for final evaluation by the experts meetings. A "final task force" meeting reviewed the health risk assessments and public and peer review comments and, where appropriate, decided upon guideline values. During preparation of the third edition of the GDWQ, it was decided to include a public review via the world wide web in the process of development of the health criteria documents.

During the preparation of health criteria documents and at experts meetings, careful consideration was given to information available in previous risk assessments carried out by the International Programme on Chemical Safety, in its Environmental Health Criteria

monographs and Concise International Chemical Assessment Documents, the International Agency for Research on Cancer, the joint FAO/WHO Meetings on Pesticide Residues and the joint FAO/WHO Expert Committee on Food Additives (which evaluates contaminants such as lead, cadmium, nitrate and nitrite in addition to food additives).

Further up-to-date information on the GDWQ and the process of their development is available on the WHO internet site and in the current edition of the GDWQ.

**Acknowledgements**

The first draft of Aldrin and Dieldrin in Drinking-water, Background document for development of WHO *Guidelines for Drinking-water Quality*, was prepared by Dr P. Toft, Canada, to whom special thanks are due.

The work of the following working group coordinators was crucial in the development of this document and others in the third edition:

Mr J.K. Fawell, United Kingdom (*Organic and inorganic constituents*)
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Prof. Y. Magara, Hokkaido University, Japan (*Analytical achievability*)
Mr P. Jackson, WRc-NSF, United Kingdom (*Treatment achievability*)

The contribution of peer reviewers is greatly appreciated. The draft text was posted on the world wide web for comments from the public. The revised text and the comments were discussed at the Final Task Force Meeting for the third edition of the GDWQ, held on 31 March to 4 April 2003, at which time the present version was finalized. The input of those who provided comments and of participants in the meeting is gratefully reflected in the final text.

The WHO coordinators were as follows:

Dr J. Bartram, Coordinator, Water Sanitation and Health Programme, WHO Headquarters, and formerly WHO European Centre for Environmental Health
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Ms C. Vickers provided a liaison with the International Chemical Safety Programme, WHO Headquarters.

Ms Marla Sheffer of Ottawa, Canada, was responsible for the scientific editing of the document.

Many individuals from various countries contributed to the development of the GDWQ. The efforts of all who contributed to the preparation of this document and in particular those who provided peer or public domain review comment are greatly appreciated.

## Acronyms and abbreviations used in the text

| | |
|---|---|
| ADI | acceptable daily intake |
| CAS | Chemical Abstracts Service |
| EPA | Environmental Protection Agency (USA) |
| FAO | Food and Agriculture Organization of the United Nations |
| HEOD | (1R,4S,4aS,5R,6R,7S,8S,8aR)-1,2,3,4,10,10-hexachloro-1,4,4a,5,6,7,8,8a-octahydro-6,7-epoxy-1,4:5,8-dimethanonaphthalene |
| HHDN | (1R,4S,4aS,5S,8R,8aR)-1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene |
| IARC | International Agency for Research on Cancer |
| IUPAC | International Union of Pure and Applied Chemistry |
| JMPR | Joint FAO/WHO Meeting on Pesticide Residues |
| $LD_{50}$ | median lethal dose |
| LOAEL | lowest-observed-adverse-effect level |
| NOAEL | no-observed-adverse-effect level |
| PTDI | provisional tolerable daily intake |
| TDI | tolerable daily intake |
| UNEP | United Nations Environment Programme |
| USA | United States of America |
| WHO | World Health Organization |

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## *1. GENERAL DESCRIPTION*

### *1.1 Identity*

| *Compound* | *CAS No.* | *Molecular formula* |
|---|---|---|
| Aldrin | 309-00-2 | $C_{12}H_8Cl_6$ |
| Dieldrin | 60-57-1 | $C_{12}H_8Cl_6O$ |

The IUPAC name for aldrin is (1R,4S,4aS,5S,8R,8aR)-1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene (HHDN). Aldrin is most commonly used to mean HHDN with a purity greater than 95%, except in Denmark and the countries of the former Soviet Union, where it is the name given to pure HHDN. Impurities include octachlorocyclopentene, hexachlorobutadiene, toluene and polymerization products (WHO, 1989).

The IUPAC name for dieldrin is (1R,4S,4aS,5R,6R,7S,8S,8aR)-1,2,3,4,10,10-hexachloro-1,4,4a,5,6,7,8,8a-octahydro-6,7-epoxy-1,4:5,8-dimethanonaphthalene (HEOD). Dieldrin is most commonly used to mean HEOD with a purity greater than 85%, except in Denmark and the countries of the former Soviet Union, where it is the name given to pure HEOD. Impurities include other polychloroepoxyocta-hydrodimethanonaphthalenes and endrin (WHO, 1989).

### *1.2 Physicochemical properties (WHO, 1989; Worthing, 1991)*

| *Property* | *Technical aldrin (95% pure)* | *Technical dieldrin* |
|---|---|---|
| Melting point (°C) | 49–60 | 175–176 |
| Density at 20 °C (g/cm$^3$) | 1.54 | 1.62 |
| Water solubility at 20 °C (µg/litre) | 27 | 186 |
| Log octanol–water partition coefficient | 3.0 | 4.6 |
| Vapour pressure at 20 °C (Pa) | $8.6 \times 10^{-3}$ | $0.4 \times 10^{-3}$ |

### *1.3 Organoleptic properties*

Odour threshold values of 17 and 41 µg/litre have been reported for aldrin and dieldrin, respectively (US EPA, 1987; Waggot & Bell, 1988).

### *1.4 Major uses*

Aldrin and dieldrin are highly effective insecticides for soil-dwelling pests and for the protection of wooden structures against termites and wood borers. Dieldrin has also been used against insects of public health importance (WHO, 1989). Although the use of aldrin and dieldrin has been severely restricted or banned in many parts of the world since the early 1970s, the insecticides are still used in termite control in some countries (Meister, 1989).

Aldrin and dieldrin were designated as persistent organic pollutants in 1997 by the Governing Council of the United Nations Environment Programme (UNEP, 1997).

### *1.5 Environmental fate*

In soil, aldrin is removed by oxidation to dieldrin and evaporation. In temperate climates, only 75% is oxidized within a year after application. The further disappearance of dieldrin is very slow under these conditions; the half-life is approximately 5 years. Under tropical conditions, both oxidation and further disappearance of dieldrin are rapid, 90% disappearing within 1 month, primarily by volatilization (WHO, 1989).

## *2. ANALYTICAL METHODS*

Aldrin and dieldrin are determined by extraction with pentane followed by gas chromatography with electron capture detection. The detection limits in tap water and river water are about 0.001 µg/litre for aldrin and 0.002 µg/litre for dieldrin.

## *3. ENVIRONMENTAL LEVELS AND HUMAN EXPOSURE*

### *3.1 Air*

Dieldrin has been detected at very low concentrations in ambient air, on dust particles and in rainwater. In non-agricultural areas, concentrations of 0.06–1.6 ng/m$^3$ have been reported; in agricultural areas, mean levels are in the range 1–2 ng/m$^3$, with a maximum of about 40 ng/m$^3$ (WHO, 1989).

Concentrations found in the air of houses treated for termites are much higher (40–7000 ng/m$^3$). The presence of aldrin/dieldrin-treated wood in houses results in indoor air concentrations of 10–500 ng/m$^3$ (WHO, 1989).

### *3.2 Water*

The concentrations of aldrin and dieldrin in aquatic environments and drinking-water are normally less than 10 ng/litre. Higher levels are attributed to contamination from industrial effluents and soil erosion during agricultural use. River sediments may contain higher amounts (up to 1 mg/kg). These pesticides are rarely present in groundwater, as little leaching from soils occurs (WHO, 1989).

### *3.3 Food*

Dieldrin is stored in the adipose tissue, liver, brain and muscle of mammals, fish, birds and other parts of the food-chain. The reduction in use since the 1970s has decreased the residues in food in many countries to well below the levels that may result in an intake of 0.1 µg/kg of body weight per day (the PTDI established by JMPR in 1994)

(FAO/WHO, 1995). The intake in 1980–1982 was estimated to be below 0.2 μg/kg of body weight per day in several countries (WHO, 1989).

Dieldrin has been detected in breast milk at a mean concentration of 0.5–11 μg/kg of milk in Europe and the USA. Breast-fed babies receive doses of approximately 1 μg/kg of body weight per day when mothers' milk contains 6 μg of dieldrin per litre (WHO, 1989). Although concentrations in breast milk decreased from an average of 1.33 μg/kg of milk in 1982 to 0.85 μg/kg of milk in 1986 (National Board of Health, 1987), higher concentrations (mean 13 μg/litre) have been found in breast milk from women whose houses were treated annually with aldrin (Stacey & Tatum, 1985).

## *4. KINETICS AND METABOLISM IN LABORATORY ANIMALS AND HUMANS*

Aldrin and dieldrin are absorbed by the oral, inhalation and dermal routes. They tend to accumulate in adipose tissue. A steady state between intake, storage and excretion is reached following repeated dosing. Aldrin and dieldrin can be mobilized from the adipose tissue compartment, causing an increase in blood level that results in toxic manifestations. Dieldrin is metabolized in the liver and is excreted, with its metabolites, primarily in the faeces via the bile in humans and in most animals tested (mouse, rat, monkey). The major metabolite is 9-hydroxy dieldrin. Small amounts of *trans*-6,7-hydroxy dieldrin, dicarboxylic acids and bridged pentachloroketone are excreted, but only in laboratory animals. The ratios between the amounts of the various metabolites produced differ for different animals (WHO, 1989).

## *5. EFFECTS ON LABORATORY ANIMALS AND IN VITRO TEST SYSTEMS*

### *5.1 Acute exposure*

Acute oral $LD_{50}$s of 33–65 mg/kg of body weight have been reported for aldrin and dieldrin for mice, rats, dogs, pigs and rabbits. The reported value for dieldrin in monkeys is 3 mg/kg of body weight (WHO, 1989).

### *5.2 Short-term exposure*

Short-term studies on rodents have shown that the liver is the major target organ of aldrin and dieldrin exposure. The liver to body weight ratio increases, and histopathological changes are observed, which have become known as "chlorinated hydrocarbon insecticide rodent liver." In rats, the changes were minimal at a dose of 0.025 mg/kg of body weight per day, and this value was selected as the LOAEL (WHO, 1989).

### *5.3 Long-term exposure*

Dogs seem more sensitive to aldrin and dieldrin than rats. In a 2-year study with beagle dogs receiving dieldrin in olive oil at doses of 0.005 or 0.05 mg/kg of body weight per day, female dogs given 0.05 mg/kg of body weight per day had an increased liver to body

weight ratio. The NOAEL was estimated to be 0.005 mg/kg of body weight per day (WHO, 1989).

### *5.4 Reproductive and developmental toxicity*

The results of a number of reproductive studies suggest that dieldrin at levels of 2 mg/kg in the rat diet and 3 mg/kg in the mouse diet has no effects on reproduction. At these levels, however, there may be biochemical and histopathological effects.

In a limited study with dogs fed aldrin or dieldrin, pup survival was generally lower. No effects were observed in dogs receiving 0.2 mg of dieldrin per kg in the diet (WHO, 1989).

### *5.5 Mutagenicity and related end-points*

The majority of studies on aldrin and dieldrin have not shown them to be mutagenic. In one study in which dieldrin was mutagenic in two out of three strains of *Salmonella typhimurium*, a dose–response relationship was not demonstrated (WHO, 1989).

### *5.6 Carcinogenicity*

A number of long-term studies have shown aldrin and dieldrin to produce benign and malignant tumours of the liver in various strains of mice but not in other species. This indicates that the effect of aldrin/dieldrin on the mouse liver is species specific. Aldrin and dieldrin have also been tested for carcinogenicity by the oral route in hamsters, dogs and monkeys (WHO, 1989). After assessing much of the available data, IARC (1987) concluded that the evidence for the carcinogenicity to animals for both aldrin and dieldrin is limited and classified both chemicals in Group 3.

## *6. EFFECTS ON HUMANS*

Both aldrin and dieldrin are highly toxic to humans, the target organs being the central nervous system and the liver. Severe cases of both accidental and occupational poisoning and a number of fatalities have been reported. The lethal dose of dieldrin is estimated to be approximately 10 mg/kg of body weight per day. The majority of those poisoned by aldrin or dieldrin recover, and irreversible effects have not been reported.

Male volunteers exposed to dieldrin doses below 3 μg/kg of body weight per day for 18 months showed no effects on health. The concentration of dieldrin in blood and adipose tissue was found to be proportional to the daily intake (WHO, 1989).

Effects on occupationally exposed workers have been studied in two epidemiological mortality studies. In one study (232 subjects), no indication of specific carcinogenic activity was found. In another study (1040 subjects), the mortality due to malignant neoplasms was lower than expected. There was a slight excess of cancers of the

oesophagus, rectum and liver, based on very small numbers. The only disease showing higher mortality rates than expected was non-malignant respiratory system disease, specifically pneumonia (WHO, 1989).

Chromosome studies have been carried out on human peripheral lymphocytes from agricultural workers and workers engaged in the control of Chagas disease with at least 10 years of exposure to dieldrin. There were no differences between the control and exposure groups in structural chromosomal aberrations and sister chromatid exchange (WHO, 1989).

## *7. GUIDELINE VALUE*

As already mentioned, IARC (1987) has classified aldrin and dieldrin in Group 3. All the available information on aldrin and dieldrin taken together, including studies on humans, supports the view that these chemicals make very little contribution, if any, to the incidence of cancer in humans. Therefore, a TDI approach can be used to calculate a guideline value.

In 1977, JMPR recommended an ADI of 0.1 µg/kg of body weight (combined total for aldrin and dieldrin). This was based on NOAELs of 1 mg/kg of diet in the dog and 0.5 mg/kg of diet in the rat, which are equivalent to 0.025 mg/kg of body weight per day in both species. JMPR applied an uncertainty factor of 250 based on concern about carcinogenicity observed in mice (FAO/WHO, 1978).

This ADI was reaffirmed by JMPR in 1994 and converted into a PTDI because aldrin and dieldrin were no longer used as pesticides (FAO/WHO, 1995). The PTDI of 0.1 µg/kg of body weight (combined total for aldrin and dieldrin) was used as the basis for the drinking-water guideline. Although levels of aldrin and dieldrin in food have been decreasing, dieldrin is highly persistent and bioaccumulates. The guideline value is therefore based on an allocation of 1% of the PTDI to drinking-water, giving a value of 0.03 µg/litre.

## *8. REFERENCES*

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