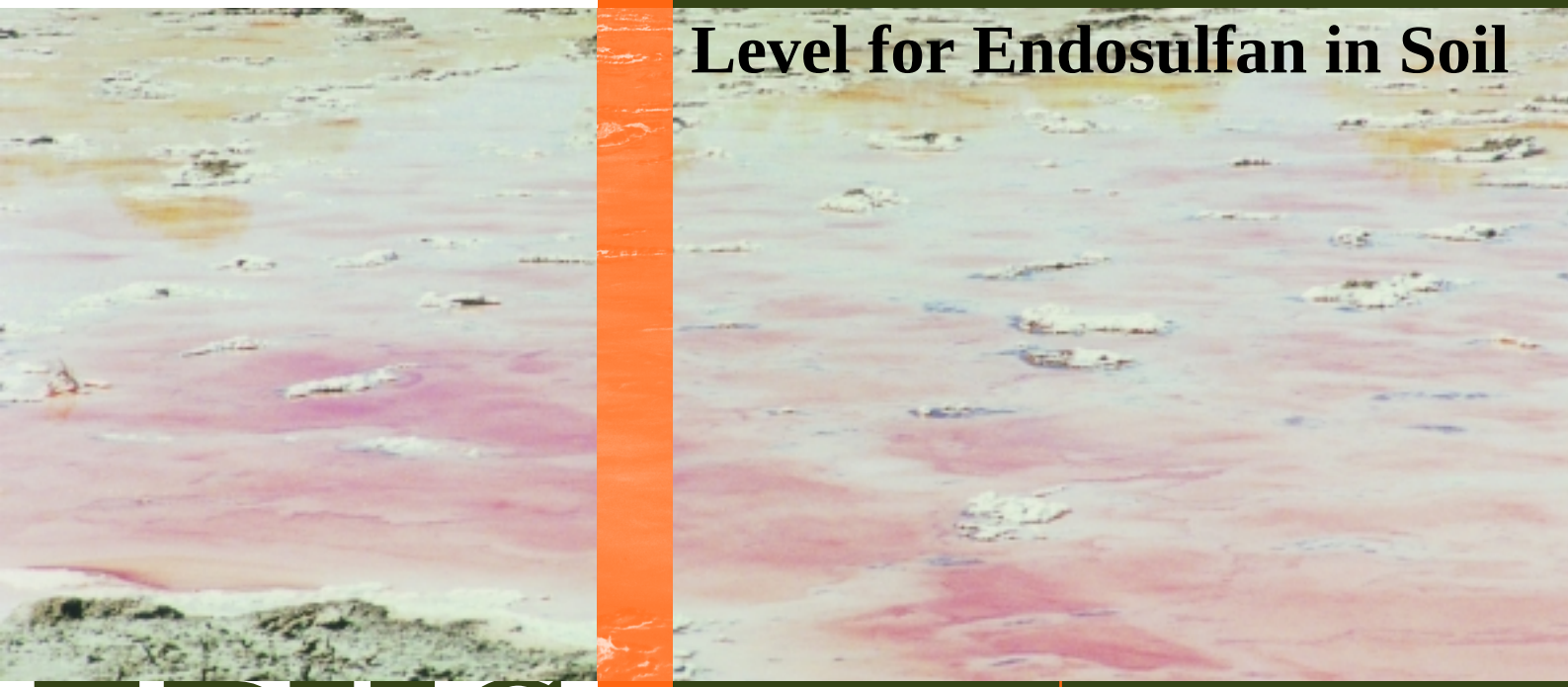
Health-Based Investigation Level for Endosulfan in Soil



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Health Investigation Level for Endosulfan in Soil

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1 INTRODUCTION

Endosulfan is a chlorinated cyclodiene pesticide predominantly used in Australia in the cotton industry. Endosulfan consists of two isomers (α and β). Labelling changes and cancellation of the use of ultra low volume formulations by the National Registration Authority (NRA) since 1999 have restricted its use in the cotton industry.

This paper describes the chemical and physical properties of endosulfan, its uses in Australia and its occurrence and distribution in the environment. It reviews the toxicology of endosulfan relevant to establishing the acceptable daily intake (ADI) and considers the potential for human exposure to occur. A HIL for endosulfan in soil is derived.

2 PROPERTIES AND USES

2.1 PHYSICAL AND CHEMICAL PROPERTIES

Endosulfan consists of the isomers α-endosulfan and β-endosulfan. The α and β isomers are present in the ratio of 7:3, respectively. Endosulfan sulfate is a reaction product found in technical endosulfan and is also found in the environment as a result of the degradation of endosulfan.

The physical and chemical properties of endosulfan, the α and β isomers and its primary metabolite are:

	Endosulfan	**α-endosulfan**	**β-endosulfan**	**Endosulfan sulfate**
Empirical formula	$C_9H_6Cl_6O_3S$	$C_9H_6Cl_6O_3S$	$C_9H_6Cl_6O_3S$	$C_9H_6Cl_6O_4S$
Molecular weight	406.95	406.93	406.93	422.9
Solubility in water (25° C) µg/L	60 to 100	530	280	117-220
Partition coefficient K_{ow}	3.55	3.83	3.52	3.66
Vapour pressure (25° C) mmHg	1×10^{-5}	1×10^{-5}	1×10^{-5}	1×10^{-5}

Source: ATSDR (2000)

2.2 USES

Endosulfan is a broad spectrum agaricide/insecticide which has been used in Australia for over 30 years, particularly in the cotton industry. It is registered in Australia for control of a large variety of insects in horticultural and agricultural crops such as citrus fruits, grains, nuts, tobacco and vegetables and is also used as a wood preservative (NRA, 1998).

3 EXPOSURE STANDARDS

3.1 ENVIRONMENTAL EXPOSURE

ANZECC guideline value of 0.01 µg/L for protection of aquatic life
ADWG guideline value of 0.00005 mg/L (based on analytical level of detection)
ADWG health value of 0.03 mg/L (based on 10% of ADI)

3.2 OCCUPATIONAL EXPOSURE

NOHSC occupational guideline of 0.1 mg/m^3 TWA

3.3 OTHER APPLICABLE GUIDELINES AND REGULATIONS

ADI	National Registration Authority	0.006 mg/kg bw/day
ADI	World Health Organization	0.006 mg/kg bw/day
RfD	US EPA	0.006 mg/kg bw/day

Australian maximum residue levels (MRLs) for food range from 0.05 to 2 mg/kg with an MRL of 30 mg/kg for tea (ANZFA, 2001).

4 ENVIRONMENTAL BEHAVIOUR, OCCURRENCE AND DISTRIBUTION

4.1 SOIL

α- and β-endosulfan and its metabolites are not very mobile in soil with most being retained in the surface 10 cm. Leaching studies indicate that less than 2% of the applied dose was recovered from leachate (NRA, 1998).

Endosulfan released to soil is most likely subject to photolysis (on soil surfaces), hydrolysis (under alkaline conditions) or biodegradation (ATSDR, 2000). The soil half-lives for the parent isomers are dependent on soil conditions and have been reported to range from 1 to 2 weeks to 2 months, but for total residues the half-lives are reported to range from 9 months to two years. Endosulfan, particularly the α-isomer, undergoes significant degradation in moist alkaline soils within a week. Degradation occurs less readily under dry anaerobic conditions (NRA, 1998). In dry soils, sulfate formation can be suppressed and under anaerobic conditions half-lives of the parent isomers and sulfate metabolite are similar.

4.2 WATER

Contamination of surface water catchments is largely attributed to run-off from sprayed areas and to a lesser extent as a result of spray drift (NRA, 1998). Given the low potential for residues to leach from soil, contamination of ground water is not expected. Parent isomers of endosulfan have low water solubility. The sulfate metabolite is more water soluble than endosulfan. The parent isomers are more water soluble than both endosulfan and endosulfan sulfate.

4.3 AIR

Significant amounts of endosulfan volatilise from soil and leaf surfaces shortly after application.

4.4 PLANTS AND OTHER DIETARY SOURCES

The 19th Australian Dietary Food Survey (ANZFA, 2001) indicates that total endosulfan residues were detected in a range of foods. The maximum level of residues found in surveyed food ranged from 0.01 mg/kg in pears, potatoes and grapes to 6.1 mg/kg in lettuce.

Metabolism studies indicate that endosulfan sulfate is the major residue in plant and animal tissues. Endosulfan sulfate has a significantly longer half-life than the parent isomers and is usually the only residue detected in animal fat (NRA, 1998).

5 ABSORPTION, DISTRIBUTION, METABOLISM AND EXCRETION

In animal studies, approximately 80% of an oral dose was absorbed from the gastrointestinal tract. Dermal absorption has been reported after occupational exposure. In animals, dermal exposure for 168 hours resulted in approximately half the applied dose being absorbed. Very few studies have examined absorption after inhalation. However, neurological effects have been observed in rats following inhalational exposure (NRA, 1998). Animal studies indicate that endosulfan can be metabolised to lipophilic compounds that can rapidly enter tissues, predominantly liver and kidney, or to more hydrophilic compounds that can be more readily excreted. In humans and animals, elimination half-lives are dependent on the dose and route of exposure and reported values range from a few days to a few weeks. Elimination of endosulfan from the body occurs mainly via the faeces and, to a lesser extent, urine (ATDSR, 2000).

Endosulfan has been found in breast milk of women environmentally exposed and in fatty tissue of children in agricultural areas (assumed to be from dietary exposure) (ATDSR, 2000).

Biological monitoring for endosulfan in body tissues and fluids will only establish whether exposure to endosulfan has recently occurred as it is readily eliminated from the body. Monitoring of the level of endosulfan in urine may be useful in occupational settings where regular exposure to endosulfan is more likely to occur.

6 TOXICOLOGY

Endosulfan is a chlorinated cyclodiene pesticide with a predominant toxicological effect following acute exposure in humans and animals being over stimulation of the central nervous system. The α-isomer is reported to be more toxic than the β-isomer (ATDSR, 2000). Signs of neurotoxicity reported in humans and animals include hyperactivity, involuntary muscle movements, pronounced sensitivity to noise and light, seizures and convulsions.

Limited information is available on the toxicity of endosulfan sulfate.

Comprehensive reviews of the toxicology of endosulfan have been undertaken by the NRA, the United States Environmental Protection Agency, and the United States Department of Health and Human Services and were reviewed for the purpose of establishing a HIL for endosulfan in soil.

For the purpose of this discussion, toxicological considerations will be limited to health effects reported in humans and toxicological studies considered by various agencies in setting the ADI for endosulfan.

6.1 HUMANS

Limited information is available on the potential for endosulfan to produce toxicity in humans in non occupational settings. Case reports of oral exposure to endosulfan resulting from accidents or self harm indicate that endosulfan has potential to produce nausea, vomiting, headaches, dizziness, tonic-clonic convulsions, unconsciousness, renal failure and liver dysfunction. According to Lehr, 1996 (cited in NRA, 1998), the lowest reported dose that resulted in death of a human was 34 mg/kg bw.

Health effects reported from exposure to endosulfan in occupational settings include systemic toxicity, eye and skin irritation, respiratory effects and headaches (NRA, 1998). No information was available on the levels of exposure required to produce these effects.

6.2 ACCEPTABLE DAILY INTAKE

An ADI of 0.006 mg/kg/day has been set for endosulfan by a number of regulatory agencies. The ADI was established on the basis of a no observed effect level (NOEL) of 0.6 mg/kg/day and the application of an uncertainty factor of 100 to account for inter- and intra-species variability. The NOEL was identified from studies undertaken in a range of species (mouse, rat and dog) and reflects the type of adverse effects that can occur following dietary exposure to endosulfan for periods ranging from 13 weeks to 2 years in rats and up to 78 weeks in mice and 1-year in dogs. Table 1 shows the no observed effect levels (NOEL), lowest observed effect levels (LOEL) and adverse effects reported for different studies that formed the basis for establishing the ADI.

Table 1: Summary of studies used for the derivation of an ADI for endosulfan

Species	Type of study	NOEL (mg/kg/day)	LOEL (mg/kg/day)	Adverse effect reported
Mouse	78-week dietary	0.58	Not determined	No effects reported at highest dose
Rat	13-week dietary	0.64	1.92	Haematological changes and granular pigment formation in the renal proximal tubules
	Developmental	0.66(f)	2.0	Decreased female (f) bodyweights
	2-year dietary	0.6	2.9	Reduced bodyweight gain and kidney pathology (reported as enlarged kidneys, marked progressive glomerulonephritis and aneurysms)
Dog	52-week dietary	0.65 (m) 0.57 (f)	1.75	Decreased bodyweight gain, and increased neurological findings (loss or weakening of placing and righting reactions, tonic contractions of abdominal muscle and masticatory muscles a few hours after feeding)

The critical study considered relevant to establishing an ADI for endosulfan is the 12-month dietary dog study. In this study, significant neurotoxicity was reported at the LOEL (1.75 mg/kg/day) and a NOEL of 0.61 mg/kg/day (average male and female) was identified. Although the duration of the study was only 12 months, results from other studies suggest that the NOEL for endosulfan for chronic toxicity was approximately 0.6 mg/kg/day.

Dietary studies conducted in rats have shown that endosulfan also has potential to effect gonadal function in male rats following doses as low as 2.5 mg/kg/day for 60 to 70 days, increase uterine and pituitary weights in first generation and second generation pups exposed to 6.18 mg/kg/day, and increase parathyroid weights in male rats exposed to 21 mg/kg/day for 2 years. These observed effects occurred at levels higher than the NOEL used in the derivation of the ADI (Table 1).

7 BACKGROUND EXPOSURE

In assessing the potential for exposure to endosulfan the main exposure pathways considered were the ingestion of residues in the diet, in water and the inhalation of endosulfan in ambient air. Limited information is available on the level of endosulfan in water and air. Information that is available is highly dependent on when sampling was undertaken in relation to when and how endosulfan was applied and farming practices.

7.1 FOOD

Based on results from the 19th Australian Dietary Food Survey (ANZFA, 2001) the Australia New Zealand Food Authority has estimated that dietary residues of endosulfan in foods contribute 1.1% of the ADI for a 2-year old. This level is comparable to the contribution of dietary residues to the ADI for 12 year olds (1.1% (m) and 0.9% (f)) and adults (1% (m) and 1% (f)).

7.2 WATER

Endosulfan is very rarely detected in Brisbane reticulated water supplies (Hodge, Queensland Health Scientific Services, pers comm). However, levels of 0.02 to 0.06 µg/L have been reported in rainwater tanks in regional Queensland. In cotton growing areas in Queensland, a mean total endosulfan level of 0.13 µg/L (maximum of 0.25 µg/L) was reported for untreated water and a mean level of 0.05 µg/L (maximum of 0.12 µg/L) after water treatment.

Levels reported in the Central and Northwest regions of NSW for the period 1991 to 2000 were well below the health based ADWG with levels not exceeding 0.6 µg/L in all river basins reported (NSW Department of Land and Conservation, 2001).

7.3 AIR

A study of pesticide exposure in the cotton growing town of Emerald in Queensland indicated that a high proportion of urban samples (approx. 75%) and samples taken near cotton fields (64%) did not contain detectable levels of endosulfan. Geometric mean values ranged from 0.015 µg/m^3 to 0.30 µg/m^3 with a maximum level of 0.9 µg/m^3 attained at a sampling site close to the cotton fields. At an exposure level of 0.9 µg/m^3, it

was estimated that for a child this level of exposure would contribute 5.6% to the ADI (Qld Health, 1991).

8 DERIVATION OF HEALTH INVESTIGATION GUIDELINE VALUE

As young children are considered the most likely to encounter significant residues of endosulfan in soil in a residential setting, the HIL for endosulfan accounts for the exposure that a 2-year old could potentially experience in such a setting. The HIL for endosulfan in soil is considered conservative, as it is unlikely that a person's daily intake of imidacloprid from soil would be maintained over a lifetime at the level estimated for a 2-year old.

For the purpose of establishing a HIL it will be assumed that the toxicological profile for endosulfan sulfate is similar to endosulfan and accordingly the HIL should be compared with the sum of endosulfan sulfate and parent isomer levels detected in the soil.

8.1 RECOMMENDED SOIL INVESTIGATION GUIDELINE FOR ENDOSULFAN

The derivation of the HIL is based on the following:

ADI	0.06 mg/kg bw/day
Bioavailability from soil	100%
Amount of soil ingested	100 mg/day

The amount of soil ingested was based on a default soil ingestion value for a 2-year old (ANZECC & NHMRC, 1992).

For a 2-year old the estimated background exposure to endosulfan from residues in air, food and water was found to be contribute approximately 7% to the ADI. As limited data were available to determine the background exposure to endosulfan it is considered reasonable that exposure to endosulfan in soil should not contribute more than 20% of the ADI.

For a 2-year old weighing 13.2 kg, an intake of endosulfan at 20% of the ADI (0.006 mg/kg bw/d) would equate to a daily intake of endosulfan of 0.01584 mg. If a 2-year old were to ingest 100 mg of soil the concentration in soil required to achieve an intake of 0.01584 mg would be 158.4 mg/kg. Accordingly, the HIL established for endosulfan and its residues in soil is **160 mg/kg**.

8.2 COMMON SOIL CONCENTRATIONS

Australian data reviewed by the NRA (NRA, 1999) indicates that soil residues largely dissipate between seasons, with sulfate residues most persistent with values in the 100-200 µg/kg range. A value of 620 µg/kg was found in the surface 10cm of soil at Narrabri, after a single application 12 months previously (only known application in 2 years).

9 FURTHER RESEARCH

Further research may be required to more comprehensively assess the toxicity of endosulfan sulfate. Derivation of background exposures was limited by available data and this may require further research. Endosulfan use in Australia has declined since the introduction of stricter labelling requirements and the ban of ultra low volume

formulations. This latter restriction should reduce the potential for background air exposure and reduce the contamination of surface water from spray drift. The decline in endosulfan use and expected associated decline in occurrence in the environment should be monitored.

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