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Canadian Soil Quality Guidelines for

Benzo(a)pyrene: Environmental

Supporting Document — Final Draft December 1996

NOTICE

This final draft document provides the information supporting the derivation of environmental soil quality guidelines for benzo(a)pyrene. Development of these soil quality guidelines was initiated through the National Contaminated Sites Remediation Program (NCSRP) which officially ended in March 1995. Given the need for national soil quality guidelines for contaminated sites management and many other applications, development was pursued under the direction of the CCME Soil Quality Guidelines Task Group after the end of the NCRSP.

This document is a working document that was released shortly after the publication of "A Protocol for the Derivation of Environmental and Human Health Soil Quality Guidelines" (CCME 1996). The CCME recognizes that some refinements or changes to the Protocol may become necessary upon application and testing. If required, amendments to the Protocol will be made and the guidelines will be modified accordingly. For this reason guidelines are referred to in this document as CCME Recommended Guidelines. Readers who wish to comment or provide suggestions on the Protocol or on the guidelines presented in this document should send them to the following address:

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The values in this document are for general guidance only. They do not establish or affect legal rights or obligations. They do not establish a binding norm, or prohibit alternatives not included in the document. They are not finally determinative of the issues addressed. Decisions in any particular case will be made by applying the law and regulations on the basis of specific facts when regulations are promulgated or permits are issued.

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This document is a supporting technical document. It is available in English only. A French Abstract is given on page vii.

Ce document technique de soutien n'est disponible qu'en anglais avec un résumé en français présenté à la page vii.

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ABSTRACT

Canadian environmental quality guidelines, developed under the auspices of the Canadian Council of Ministers of the Environment (CCME), are numerical concentrations or narrative statements recommended to support and maintain designated resource uses. CCME Canadian soil quality guidelines can be used as the basis for consistent assessment and remediation of contaminants at sites in Canada.

This report was prepared by the Guidelines Division of the Science Policy and Environmental Quality Branch (Environment Canada), which acts as Technical Secretariat for the CCME Soil Quality Guidelines Task Group. The Guidelines were derived according to the procedures described in *A Protocol for the Derivation of Environmental and Human Health Soil Quality Guidelines* (CCME 1996).

Following the introduction, chapter 2 presents chemical and physical properties of benzo(a)pyrene (BaP) and a review of the sources and emissions in Canada. Chapter 3 discusses BaP's distribution and behavior in the environment while chapter 4 reports the toxicological effects of BaP on microbial processes, plants, and animals. These informations are used in chapter 5 to derive soil quality guidelines for BaP to protect environmental receptors in four types of land uses: agricultural, residential/parkland, commercial, and industrial.

The following soil quality guidelines are recommended by the CCME based on the available scientific data. For BaP, the available data were insufficient/inadequate to derive soil quality guidelines according to the CCME 1996 Protocol. Therefore, the Provisional Method, described in CCME (1997) and hereafter was used to derive provisional environmental soil quality guidelines for BaP. The provisional environmental soil quality guideline (SQG_E—provisional) for agricultural and residential/parkland land uses is 0.7 mg BaP·kg⁻¹ soil whereas for commercial and industrial land uses it is 1.4 mg BaP·kg⁻¹ soil.

RÉSUMÉ

Les recommandations canadiennes pour la qualité de l'environnement, élaborées sous les auspices du Conseil Canadien des Ministres de l'Environnement (CCME), sont des concentrations ou des énoncés décrivant les limites recommandées dans le but d'assurer le maintien et le développement durable d'utilisations désignées des ressources. Les recommandations canadiennes pour la qualité des sols proposées par le CCME peuvent être utilisées comme base pour l'uniformisation des processus d'évaluation et d'assainissement des terrains contaminés au Canada.

Le présent document a été préparé par la Division des Recommandations de la Direction de la Qualité de l'Environnement et de la Politique Scientifique (Environnement Canada), qui agit comme secrétaire technique pour le Groupe de Travail du CCME sur les Recommandation pour la qualité des sols. Les Recommandations ont été élaborées selon les procédures décrites dans le *Protocole d'élaboration de recommandations pour la qualité des sols en fonction de l'environnement et de la santé humaine* (CCME 1996).

Faisant suite à une brève introduction, le chapitre 2 présente les propriétés physiques et chimiques du benzo(a)pyrene (BaP) de même qu'un survol des sources et des émissions au Canada. Le chapitre 3 discute du devenir et du comportement de cette substance dans l'environnement alors que le chapitre 4 rapporte ses effets toxicologique sur les processus microbiens, les plantes et les animaux. Ces informations sont utilisées au chapitre 5 afin d'élaborer des recommandations pour la qualité des sols relatives au BaP en vue de la protection de l'environnement dans le cadre de quatre types d'utilisations de terrains: agricole, résidentiel/parc, commercial et industriel.

Les recommandation pour la qualité des sols suivantes, proposées par le CCME, sont fondées sur les données scientifiques disponibles. Pour le BaP, les données disponibles sont insuffisantes/inadéquates pour permettre l'élaboration de recommandations selon les procédures décrites dans le Protocole du CCME (1996). Ainsi, la Méthode provisoire, décrite dans un document du CCME (1997) et ci-après, à été utilisée pour élaborer des recommandations provisoires pour la qualité des sols relatives au BaP en vue de la protection de l'environnement. La recommandation provisoire pour la qualité des sols en vue de la protection de l'environnement (RQS_E—provisoire) relative aux terrains à vocation agricole et résidentielle/parc est de 0.7 mg de BaP·kg⁻¹ de sol, alors que pour les terrains à vocation commerciale et industrielle, elle est de 1.4 mg de BaP·kg⁻¹ de sol.

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1. INTRODUCTION

The Canadian Council of Ministers of the Environment's (CCME) Canadian Environmental Quality Guidelines are numerical limits for contaminants intended to maintain, improve, or protect environmental quality and human health. CCME Canadian Soil Quality Guidelines can be used as the basis for consistent assessment and remediation of contaminants at sites in Canada along with CCME guidelines issued for the protection of water quality, sediment quality, and tissue quality. In response to the urgent need to begin remediation of high priority "orphan" contaminated sites, an interim set of criteria was adopted from values currently in use in various jurisdictions across Canada (CCME 1991). Many of the CCME interim soil remediation criteria do not have a complete supporting scientific rationale and are being updated based on current scientific information.

This report reviews the sources and emissions of benzo(a)pyrene, its distribution and behaviour in the environment, and its toxicological effects on microbial processes, plants, and animals. This information is used to derive guidelines for benzo(a)pyrene to protect ecological receptors according to the processes outlined in CCME (1996) for agricultural, residential/parkland, commercial, and industrial land uses.

The values derived herein are environmental soil quality guidelines and are intended as general guidance. Site specific conditions should be considered in the application of these values. The values may be applied differently in various jurisdictions, therefore the reader should consult the appropriate jurisdiction before application of the values.

2. BACKGROUND INFORMATION

2.1 Physical and Chemical Properties

Benzo(a)pyrene (BaP) ($C_{20}H_{12}$) (CAS No. 58-32-8) is a five-ringed polycyclic aromatic hydrocarbon (PAH) formed during the thermal decomposition of organic matter. PAHs are organic compounds consisting of two or more fused benzene rings. One of the most potent and ubiquitous of the PAH carcinogens, BaP consitutes 1 - 20% of PAH emissions and is often used as an indicator of total PAH (Andelman and Suess 1970; Suess 1976; Allaire et al. 1993). BaP is one of the most studied polycylic aromatic hydrocarbon (PAH) compounds (Edwards 1983). In its purified form, BaP is characterized by fine, needle-like, pale yellow crystals that fluoresce yellow-green in the presence of UV light. BaP is negligibly volatile, has no odour, and usually occurs in complex mixtures containing many other PAHs, phenols, and aromatic amines (ATSDR 1990). The physical and chemical properties of BaP are listed in Table 1.

2.2 Analytical Methods

Analytical methods recommended for BaP by the CCME include Method 6410B and U.S. EPA Method 8270B, Revision 2 (CCME 1993).

Method 6410B, entitled "Liquid-Liquid Extraction Gas Chromatographic/Mass Spectrometric Method for Organic Compounds in Municipal and Industrial Discharges", is applicable to the determination of semivolatile organic compounds in discharges that are partitioned into an organic solvent and are amenable to gas chromatography. This method is suitable for the analysis of surface and groundwater but not for soils or sediments. The detection limit for BaP is 2.5 μ g·L⁻¹ with a range of 5 - 1300 μ g·L⁻¹ (CCME 1993).

U.S. EPA Method 8270B, Revision 2, entitled "Semivolatile Organic Compounds by Gas Chromatography/Mass Spectrometry (GC/MS) Capillary Column Technique", is used to determine the concentration of semivolatile organic compounds in extracts prepared from all types of solid waste matrices, soils and groundwater. This method is also applicable to surface water samples. This method is relatively complex and expensive to run but is one of the most comprehensive methods available for the analysis of organic pollutants. The detection limit for BaP is 10 μ g·L⁻¹ in groundwater and 0.66 mg·kg⁻¹ wet weight in soils and sediments (CCME 1993).

2.3 Production, Uses, and Global Sources

BaP is not used in industry but is produced commercially in small amounts by specialized chemical firms for research purposes (IARC 1983; Aldrich Chemical Company 1984). BaP occurs naturally in fossil fuels and persists as a contaminating by-product in several oil and coal derivatives. Coal tar contains approximately 30 mg BaP·kg⁻¹ while coal tar pitch contains 10 mg·kg¹ and creosote oil contains <0.01 mg BaP·kg⁻¹ (ATSDR 1990). Bitumen samples have been reported to contain from 0.1 to 27 mg BaP·kg⁻¹ (ATSDR 1990). The amount of BaP in motor oil increases significantly during motor operation, with up to 10 times as much PAH collected in motor oil as is released in exhaust per mile travelled (Peake and Parker 1980). New automobile motor oil contains levels of BaP up to 0.266 mg·kg⁻¹ while used motor oil may contain up to 216.8 mg·kg⁻¹ (Vazquez-Duhalt 1989).

Uses

Due to its carcinogenic and mutagenic properties, BaP is used in cancer research laboratories (ATSDR 1990). Although BaP itself is not used in industry, several BaP-containing products are in general use. Coal tar pitch produced by the coke industry is primarily used to bind carbon electrodes in aluminum reduction processes, and, to a lesser extent, as an adhesive in membrane roofs (McNeil 1983). BaP-containing creosote is used as a wood preservative for railway ties, telephone and telegraph poles, marine pilings, and other structures. A small amount of creosote is also used as a fuel by steel producers. Bitumens and asphalt are used for paving roads, waterproofing and roofing, electrical insulation, sound insulation and pipe coating. Coal tar is used extensively to treat various skin disorders, including eczema, dermatitis, and psoriasis (ATSDR 1990).

Natural Sources

BaP may be formed by the thermal decomposition of any organic material containing carbon and hydrogen. The two major mechanisms resulting in the formation of BaP are pyrolysis and the

carbonization process (e.g., during the formation of mineral oil and coal) (Bjørseth and Ramdahl 1985). Several natural processes contribute BaP to the environment, namely volcanoes, natural forest and prairie fires, biosynthetic processes, coal and mineral oil formation, and rotting vegetal and animal matter (Blumer 1961; Suess 1976; Edwards 1983; Uthe 1991). BaP concentrations in crude petroleum oil have been reported to be as high as 1000 μ g·L⁻¹, suggesting the synthesis of BaP by plants or the formation of BaP during the natural conversion of biomass to oil (Andelman and Suess 1970; Edwards 1983). Several species of phytoplankton, algae, plants and microbes have been reported to synthesize BaP, including lentil, rye and wheat seedlings (Gräf and Diehl 1966), the bacteria *Clostridium putride* and *Escherichia coli*, and the fresh water algae *Chlorella vulgaris* (Borneff et al. 1968; Suess 1976; Butler et al. 1984). Quantities of PAH's formed by natural processes are very small compared with those formed by anthropogenic processes (Suess 1976; Edwards 1983; Sims and Overcash 1983; Grimmer and Pott 1983).

Anthropogenic Sources

The origin of most PAH's, including BaP, in the environment is through the combustion of fossil fuels and refuse, and through agricultural burning (Edwards 1983; ATSDR 1990). Most (98%) natural and anthropogenic BaP is emitted directly into the atmosphere, while the remainder is released to water and land (Suess 1976; Eisler 1987; ATSDR 1990). Estimates of relative emission factors and of the production/consumption capacities of their sources indicate that combustion of wood, coal, and gas for residential heating, refuse and open burning (e.g., forest fires), industrial processes (mostly coke manufacturing and aluminum production), and automobile exhaust are the major sources of BaP emission in North America (Bjørseth and Ramdahl 1985; Eisler 1987). Other sources of BaP include: carbon black production, petroleum catalytic cracking, asphalt production, emissions from the ferroalloy industry, rubber tire wear, exhaust from airplanes and sea traffic, and cigarette smoke (IARC 1983; Sims and Overcash 1983; Bjørseth and Ramdahl 1985).

2.4 Levels in the Canadian Environment

Although PAHs are ubiquitous in nature, anthropogenic sources account for most of the BaP in the global environment (Eisler 1987). Until the beginning of this century, a natural balance existed between the production and the degradation of PAHs; however, increased industrialization and the rising rate of consumption of wood and fossil fuels has increased the rate of formation of PAHs while their rate of decomposition has remained constant (Suess 1976). Detectable levels of BaP are common in Canadian soils, water, and atmosphere, particularly near emission sources.

Air

Annual global emissions of BaP have been estimated at 4 600 tonnes during 1966 - 1969 (Suess 1976) and at 5 000 tonnes in the mid-seventies and at present time (Eisler 1987; Uthe 1991). An estimated 6 000 metric tonnes of PAH, including more than 1 200 tonnes of BaP, are released into the atmosphere annually in the United States (Grimmer and Pott 1983; Bjørseth and Ramdahl 1985; Eisler 1987). Estimates of atmospheric emissions of BaP in the U.S. include 588 tonnes from refuse

and open burning, 475 tonnes from heating and power sources, 198 tonnes from industrial processes (mostly coke production), and 22 tonnes from motor vehicles (Eisler 1987).

Total annual BaP emissions have been estimated at 5.1 tonnes in Ontario and 340 tonnes in Eastern North America (Ortech 1992). The most significant sources of PAH's in Ontario are residential fuelwood combustion (140 tonnes per year), ferroalloy production (53.5 tonnes per year), metallurgical coke production (26.2 tonnes per year), emissions from gasoline and diesel fuel vehicles (24.5 tonnes per year), open fires and prescribed burning (17.4 tonnes per year), and electrical power generation (8.8 tonnes per year).

Battery stack emissions of BaP from coke pushing operations at a Stelco plant were estimated to be 1.4 mg BaP·tonne⁻¹ of coal, and PAH emission rates from two Ontario Hydro thermal generating stations at Lambton and Nanticoke were $3.4 \ \mu g \cdot s^{-1}$ and $115 \ \mu g \cdot s^{-1}$, respectively (OMOE 1992). BaP emissions from the open combustion of scrap tires in a controlled experiment ranged from 84.8 - 113.9 mg BaP per kg tire (Lemieux and Ryan 1993). Jet aircrafts have emission rates of 2 - 4 mg \cdot min⁻¹ when cruising and up to 40 000 mg min during take-off (Shabad 1980). Aluminum smelting accounts for 72% of total BaP emissions (estimated at 73.2 tonnes per year) in Quebec (Dann 1989). Regular monitoring of an aluminum smelting plant at Jonquière, Quebec showed emissions of approximately 12 tonnes of BaP in 1990, a reduction of 67% since 1984 (Allaire et al. 1993).

Most PAHs in air are associated with particulates, especially soot, and PAH deposition is characteristic of particulate deposition (Pierce and Katz 1975; ATSDR 1990; Uthe 1991). The range of BaP in unpolluted non-urban air has been reported to vary between 0.1 - 0.5 ng·m⁻³ (Suess 1976). Rural air samples from Walpole Island (Ontario) in 1988 - 1989 showed mean BaP concentrations of 0.08 ng·m⁻³ (maximum 0.70 ng·m³) (Ringuette et al. 1993) while air samples collected from Barrow, Alaska, a remote Arctic site, contained mean BaP concentrations of 30 ng·m⁻³ in March 1979 and 10 ng·m⁻³ in August 1979 (Daisey et al. 1981). These values are probably representative of the natural atmospheric background.

Air concentrations of BaP in 21 Ontario urban centres in 1961 - 1962 averaged 7.1 ng·m⁻³ in the winter and 3.49 ng·m⁻³ in the summer (Grimmer and Pott 1983). The ambient air concentration of BaP at five locations near Toronto, Ontario from 1972 - 1974 showed levels ranging from 0.11 - 0.83 ng·m⁻³, with the highest concentrations found in urban/suburban areas and the lowest found in suburban-rural and rural areas (Pierce and Katz 1975). Particle-bound BaP concentrations in Ontario in the mid-seventies showed levels ranging from 0.4 - 1.7 ng·m⁻³ in Toronto, 1.4 - 3.5 ng·m³ in Hamilton, 0.1 - 0.6 ng·m⁻³ in Sarnia and 0.1 - 0.4 ng·m⁻³ in Sudbury (OMOE 1992). Mean BaP concentrations of 42 samples from Toronto and of 29 samples from Montreal collected from 1984 - 1986 were 0.30 ng·m⁻³ (maximum 3.14 ng·m⁻³) and 0.64 ng·m⁻³ (maximum 4.09 ng·m⁻³) respectively (Dann 1989). Air samples collected in the vicinity of a steel mill in northern Ontario from 1971 - 1979 had mean BaP levels of 9.4 ng·m⁻³ (maximum 110 ng·m⁻³) at a distance of 0.8 km, and 0.4 ng·m⁻³ (maximum 7.9 ng·m³) at a distance of 2.8 km (Potvin et al. 1981). Niagara River air samples collected in January 1983 showed atmospheric particulate BaP concentrations of 0.230 ± 0.440 ng·m⁻³; considerable temporal variability was observed over the two-week sampling period suggesting

local episodic emissions (Hoff and Chan 1987). Ambient air concentrations of BaP in the vicinity of aluminum smelters in Kitimat, Shawinigan and Jonquière (1989 - 1991) were 2.06, 28.47 and 36.37 ng·m⁻³ respectively (Ringuette et al. 1993). Air sampling stations within 6 km of an aluminum smelting plant at Jonquière, Quebec reported mean ambient BaP levels of 1 - 3 ng·m⁻³ in 1990 (Allaire et al. 1993).

Soil

The major source of soil contamination by PAH's is wet and dry deposition from air (Suess 1976). Secondary sources include gasoline spills, coal tar, coal tar pitch, shale oil, carbon black, cracked mineral oil effluents from various industrial processes, surface runoff and sewage sludge used as fertilizer (Andelman and Suess 1970; Edwards 1983; OMOE 1992). Atmospheric deposition accounts for 75% of the annual PAH loading to Ontario ecosystems, while land treatment with sewage sludge represents the most important direct source of PAHs to soils (OMOE 1992). Although they contribute only small amounts of BaP to the environment on a global scale, industrial and municipal waste products and hazardous waste sites may be concentrated sources of BaP to soils on a local scale (Edwards 1983; ATSDR 1990).

PAH levels in precipitation are significantly higher in winter than in summer, primarily due to increased emissions from household heating (Eisler 1987). BaP emissions and deposition from jet and automobile exhaust are localized near runways and roadways, and snow, vegetation and soils near these areas may contain relatively high concentrations of BaP (Shabad et al. 1971; Edwards 1983; Butler et al. 1984; Johnston and Harrison 1984). The maximum rate of BaP deposition near the M6 motorway in Lancaster, UK was estimated at 4.9 μ g·m⁻²·week⁻¹ at a distance of 3 to 8 metres, and 0.31 μ g·m⁻²·week⁻¹ at a distance of 9 to 47 metres (Johnston and Harrison 1984). The PAH content of surface runoff in Swedish traffic zones was found to be approximately 20 - 200 μ g·L⁻¹, while runoff from residential zones was 0.4 - 4 μ g·L⁻¹ (Vazquez-Duhalt 1989). Other rates of BaP deposition and release to the soil environment are unknown.

Biosynthesis of soil bacteria and the degradation of plant material result in a natural carcinogenic PAH concentration of upper soil layers in the range of $0.1 - 1 \text{ mg} \cdot \text{kg}^{-1}$ (Andelman and Suess 1970). Soil samples from isolated areas in the USSR had BaP concentrations of approximately 0.001 mg \cdot kg⁻¹ with no samples exceeding 0.01 mg \cdot kg⁻¹. It was suggested that the natural background level of BaP in soil was in the range of 0.001 - 0.003 mg \cdot kg⁻¹ and not exceeding 0.01 mg \cdot kg⁻¹ (Shabad et al. 1971). Soil samples taken from regions of permafrost in the USSR from layers aged 10 - 10 000 years showed BaP levels ranging from 0.0023 - 0.0052 mg \cdot kg⁻¹ dry soil (Ilnitsky et al. 1979).

Ontario background concentration (OTR₉₈) for rural parkland has been established at 0.04 mg·kg⁻¹ and a provisional OTR₉₈ for old urban parkland was established at 0.81 mg·kg⁻¹ (OMEE 1993). Previously, mean urban soil BaP levels in Ontario were reported to range from non-detection (0.005 mg·kg⁻¹ detection limit) to 0.18 mg·kg⁻¹ with a mean value of 0.028 mg·kg⁻¹ (CCME 1989). Cultivated soil located 50 m from Highway 401 in Toronto receiving both airborne and road runoff contaminants showed BaP concentrations of 0.108 mg·kg⁻¹ at the soil surface, 0.065 mg·kg¹ at a depth of 5 cm, and 0.087 mg·kg⁻¹ at a depth of 15 cm (Wang and Meresz 1981). Soil analysis at the LaSalle coking factory in Quebec revealed BaP levels ranging from the detection limit to 1300 mg·kg⁻¹; following restoration work, the site now meets the Canadian Interim Soil Quality Criteria with a BaP concentration of 10 mg·kg⁻¹ (Argus Groupe Conseil Inc. 1991). Soil samples taken from Reykjavik airport were contaminated, with BaP levels reaching 0.785 mg·kg⁻¹ (Grimmer and Pott 1983); corresponding values for Canadian airports were not available.

Agricultural lands in Canada may be periodically amended with sewage sludge; 17.8% of refinery waste is applied to land while 35.7% is disposed of in landfills. The treatment of refinery wastewater results in the accumulation of PAHs, including BaP, in sewage sludge. Refinery sludge in Burlington, Ontario had individual PAH concentrations ranging from $0.7 - 1.7 \text{ mg} \cdot \text{kg}^{-1}$ (Bulman et al. 1985).

Water

Major sources of BaP to the aquatic environment include wet and dry deposition of airborne particulates, spillage of petroleum and petroleum products into water bodies, domestic and industrial sewage effluents, surface land runoff and biosynthesis. An estimated 2 700 tonnes of BaP enter the global aquatic environment annually, largely through atmospheric deposition (1 700 tonnes) and surface runoff (960 tonnes) (OMOE 1992). The annual atmospheric flux of BaP to the Great Lakes has been estimated as 7.9 tonnes to Lake Superior, 4.0 - 5.6 tonnes to Lake Michigan, 5.8 tonnes to Lake Huron, 2.5 tonnes to Lake Erie and 1.8 tonnes to Lake Ontario (Eadie et al. 1983). Effluents discharged from the Hamilton water pollution control plant in 1983 contained BaP levels between $0.0 - 2.7 \ \mu g \cdot L^{-1}$ with an average concentration of $0.62 \ \mu g \cdot L^{-1}$; this level of discharge represented an annual loading of 69.4 kg (Zukovs et al. 1984).

Ice collected from glaciers ranging in age from 1 - 120 years showed BaP concentrations of $0.001 - 0.003 \ \mu g \cdot L^{-1}$. These results were comparable with the BaP content found in the water of non-polluted surface reservoirs (Ilnitsky et al. 1979). Uncontaminated and slightly-contaminated fresh water lakes with normal aquatic biota show BaP concentrations in the range $0.010 - 0.025 \ \mu g \cdot L^{-1}$ (Andelman and Suess 1970).

BaP concentrations detected in surface water samples from one British Columbia site and 49 locations in Alberta were all less than 1 μ g·L⁻¹. BaP levels of 0.030 - 0.043 μ g·L⁻¹ have been found in the St. Lawrence River in Quebec while 25 other sites in Quebec reported concentrations at or slightly above the detection limit of 0.00046 μ g·L⁻¹. Surface water BaP levels in the Maritimes and Newfoundland ranged from 0.0002 - 0.131 μ g·L⁻¹ in Prince Edward Island (30 sites) and 0.0001 - 0.003 μ g·L⁻¹ in New Brunswick, Nova Scotia, and Newfoundland (142 sites) (ENVIRODAT 1993). Analyses of six Great Lake water samples in the early 1980s indicated BaP concentrations of approximately 0.012 μ g·L⁻¹ (Eadie et al. 1983). PAH concentrations in drinking water are rarely high (Uthe 1991). Concentrations of BaP in U.S. surface waters used as drinking water ranged between 0.0003 - 0.002 μ g·L⁻¹, while untreated water concentrations ranged between 0.0006 - 0.210 μ g·L⁻¹ (ATSDR 1990).

Sediments

Sediments collected from a remote freshwater lake in Ontario showed a BaP concentration of 13

mg·kg⁻¹; this value was comparable with sediment measurements reported off the west coast of Greenland and probably reflects natural BaP levels (Brown and Starnes 1978).

Due to its low solubility and high propensity for binding to particulate or organic matter, most BaP (88%) in aquatic systems is bound to suspended particles or bed sediments (ATSDR 1990; Broman et al. 1991; Uthe 1991) and all but the most heavily contaminated water systems show total BaP concentrations in water of the ppt or low ppb range (Eisler 1987). Localized BaP source inputs (e.g., industrial waste effluents) combined with BaP's hydrophobicity generally result in spatial variability in BaP concentrations in the aquatic environment (Eisler 1987). Concentrations tend to be highest in inshore surface sediments and decrease with distance offshore (Uthe 1991).

Measured BaP concentrations in the sediments of the Great Lakes in the early 1980s include: 0.028 $mg \cdot kg^{-1}$ in Lake Superior, 0.004 - 0.944 $mg \cdot kg^{-1}$ in Lake Michigan, 0.023 - 0.294 $mg \cdot kg^{-1}$ in Lake Huron, 0.056 - 0.173 $mg \cdot kg^{-1}$ in Lake Erie, and 0.076 - 0.306 $mg \cdot kg^{-1}$ in Lake Ontario (Eadie et al. 1983). Concentrations of BaP in the surficial sediments of Lake Superior in 1984 - 1985 were reported to range from 0.0317 - 0.0640 $mg \cdot kg^{-1}$ with a mean value of 0.0453 $mg \cdot kg^{-1}$, while suspended solids contained 0.003 - 1 $mg \cdot kg^{-1}$ (mean = 0.034 $mg \cdot kg^{-1}$) (Baker et al. 1991). Bottom sediment samples from rivers, lakes and harbours across Canada showed BaP levels ranging from non-detection (0.001 - 0.005 $mg \cdot kg^{-1}$) to 15.0 $mg \cdot kg^{-1}$; most sediments showed mean levels of BaP less than 0.20 $mg \cdot kg^{-1}$ dry weight with the exceptions of St. John's Harbour, Newfoundland (2.1 - 3.1 $mg \cdot kg^{-1}$) and Red River, Manitoba (0.16 - 0.33 $mg \cdot kg^{-1}$) (Watt et al. 1984). A detailed survey of Sydney Harbour revealed sediment BaP levels ranging from 0.0032 - 110 $mg \cdot kg^{-1}$ in 1981; the highest levels were associated with the tar pond in Muggah Creek (Matheson et al. 1983).

The surficial sediments of St. Mary's River in Ontario contained BaP levels of up to 48 mg·kg⁻¹ dry weight in 1985 (Kauss and Hamdy 1991). Concentrations of BaP in Hamilton Harbour sediments were reported to be in the range 1.2 - 11.1 mg·kg⁻¹ in 1982, with the highest concentrations found at locations closest to steel mills (Poulton 1987). Freshwater sediment samples from eight sites in Quebec were found to contain BaP concentrations of 0.03 mg·kg⁻¹, while four sites in New Brunswick reported BaP levels of 0.003 mg·kg⁻¹ (ENVIRODAT 1993). Sediments collected from 33 sites in the Trenton Channel in the Detroit River in 1986/87 contained 0.033 - 9.9 mg·kg⁻¹ dry weight of sediment; this variability resulted from complex interactions between contaminant sources and the suspended sediment load (Furlong et al. 1988). Sediments from the Bay of Quinte, Lake Ontario contained 0.10 - 1.70 mg·kg⁻¹ BaP in 1988, with the highest concentrations found at the mouth of the Trent and Moira rivers (Poulton 1992). Sediments collected since 1986 at Kane Dock, Niagara River, an area known to be contaminated with coal tar, contained 2 mg·kg⁻¹ BaP (Dickman et al. 1992).

Groundwater

Uncontaminated groundwater shows carcinogenic PAH concentrations in the range $0.001 - 0.010 \ \mu g \cdot L^{-1}$, probably as a result of leaching from overlying soil layers (Andelman and Suess 1970). Worldwide groundwater PAH levels have been estimated variously at $0.01 - 0.05 \ \mu g \cdot L^{-1}$ (Eisler 1987) and 0.045 - 0.51 μ g·L⁻¹ (Eisler 1987). Groundwater analysis in the vicinity of the Domtar Wood Preserving Plant in New Brunswick showed BaP concentrations ranging from non-detectable to 0.32 μ g·L⁻¹ (Ringuette et al. 1993). Other Canadian levels of groundwater BaP were not available.

Biota

In general, most PAH contamination of vegetation is by direct deposition from the atmosphere, resulting in higher PAH concentrations in above-ground plant parts than in below-ground parts and higher levels in broad-leaved plants than plants with narrow leaves (Shabad 1968; Edwards 1983). Concentrations of PAHs in vegetation tend to be less than the concentrations in the soil where they grow (Edwards 1983). An analysis of onions, beets and tomatoes growing near Highway 401 in Toronto showed BaP concentrations of 0.00001 - 0.00736 mg·kg⁻¹ in the peels of the vegetables with little of the compound (0.00001 - 0.00006 mg·kg⁻¹) found in the internal tissues (Wang and Meresz 1981). Vegetation near an aluminum electrolysis plant in Jonquière, Quebec showed BaP levels of 0.01374 mg·kg⁻¹ dry weight while control plants located 140 km south of the plant contained 0.00233 mg·kg⁻¹ dry weight (Blondin and Viau 1992). Oils extracted from plants appear to have higher BaP concentrations than the original plant tissues (Edwards 1983). Levels of carcinogenic PAHs in Canadian vegetable and dairy products are believed to be in the low ppb range (Uthe 1991).

Animals of various species may bioaccumulate PAHs depending on exposure, uptake and degradation rates. Many higher animals metabolize PAHs and thus do not accumulate them, while some lower animals, such as shellfish, do not metabolize PAHs and may accumulate high concentrations, especially near PAH sources (Sirota and Uthe 1981; Uthe 1991). Most data on BaP in biota involve animals from the aquatic and marine environment (Ringuette et al. 1993). Fish from Lake Ontario and Lake Superior showed maximum BaP levels of 0.0032 and 0.0049 mg·kg⁻¹ respectively (Zenon 1985). Lobsters captured nearshore in Nova Scotia contained BaP levels of 0.0002 - 0.0009 mg·kg⁻¹ wet weight, while lobsters held for a few months in a tidal storage pound partially constructed with creosote-treated lumber showed significantly higher BaP levels ranging from 0.0074 - 0.281 mg·kg⁻¹ wet weight (Uthe 1991). Two species of mussels collected from Sydney Harbour in 1981 contained BaP levels ranging from 0.0063 - 0.280 mg kg⁻¹ wet weight; the highest body burdens of BaP were found at the sites closest to the heavily polluted sediments of Muggah Creek (Matheson et al. 1983). "Clean" mussels exposed to ambient BaP levels for three weeks in St. Mary's River, Ontario accumulated 0.0002 - 0.013 mg·kg⁻¹ wet weight, likely via water filtration (Kauss and Hamdy 1991). BaP levels in the lipids of herring gulls (Larus argentatus) from Pigeon Island and Kingston, Ontario were 3.8×10^{-5} and 3.0×10^{-5} mg·kg⁻¹ respectively (OMOE 1992).

2.5 Existing Criteria and Guidelines

Guidelines and/or criteria for BaP in soils have been developed by several jurisdictions and are presented in Table 2. In general, soils containing $<0.1 \text{ mg}\cdot\text{kg}^{-1}$ BaP are considered uncontaminated, soils containing $0.1 - 1.0 \text{ mg}\cdot\text{kg}^{-1}$ BaP are considered slightly contaminated, and soils containing $1 - 10 \text{ mg}\cdot\text{kg}^{-1}$ BaP are considered to be significantly contaminated.

3. ENVIRONMENTAL FATE AND BEHAVIOUR

3.1 Soil

Much of the BaP released into the atmosphere reaches the soil environment by wet and dry deposition (Eisler 1987). As a result of its very low vapour pressure, low solubility and tendency to bind to organic matter, BaP deposited to soil is likely to remain adsorbed to soil particles (ATSDR 1990). Desorption from soil into water or air is very unlikely, although limited leaching of BaP to groundwater may occur, particularly in the presence of organic solvents (Miller et al. 1987; ATSDR 1990). The most important fate processes for BaP in soils are adsorption and biodegradation (ATSDR 1990). Adsorption plays a large role in the bioavailability of BaP, thus affecting uptake and elimination through both biological and abiotic means (ATSDR 1990). The rate of biodegradation in soils is very slow and BaP may therefore be persistent in the soil environment (ATSDR 1990). Photochemical reactions will proceed only at soil depths of less than 1 mm (Miller et al. 1989). Volatilization of BaP from soils occurs only from the fraction of the compound not adsorbed to particulates (Bulman et al. 1985) and experimental data have shown that the rate of volatilization of BaP from soils is insignificant (Park et al. 1990; Wild and Jones 1993).

Adsorption and leaching

The adsorption of BaP to soil particulates occurs predominantly through non-site specific processes such as Van der Waals's forces (Bulman et al. 1985). The adsorption and leaching of BaP in soils are complex and dependent upon many variables, including soil type, soil moisture, temperature, presence of competing molecules and solvents, and pH. The most important soil property affecting the sorption of BaP is the soil organic matter content (Sims and Overcash 1983; Bulman et al. 1985). Sorption equilibrium of PAHs to soils and sediments is generally reached within 24 hours (Bulman et al. 1985), while BaP sorption equilibrium to sewage sludge occurs in approximately 30 minutes (Morozzi and Scardazza 1988). Adsorption to soil particulates may limit the availability of BaP for photochemical and biological degradation, while the presence of organic solvents may increase the bioavailability of BaP and permit its uptake by plants (Bulman et al. 1985). Leaching of BaP with colloidal material and rapid movement down macropores account for only minimal loss from soils (Wild et al. 1990).

Biodegradation

Biodegradation is the major fate process for BaP in soils (ATSDR 1990). BaP may be degraded by bacteria, fungi, plants and, to a lesser extent, by higher animals. In general, the rate of microbial degradation is very slow and BaP is often persistent in soils. Products of microbial degradation may include benzoic acid, phthalic anhydride, dimethyl phthalate and methoxybenzopyrene (Miller et al. 1988). Soil factors affecting the rate of biodegradation of BaP include salinity, moisture, illumination, aeration, temperature, pH, redox potential, nutrient availability, soil type, soil history and BaP concentration, while microbial factors include the species present and their acclimation status (Sims and Overcash 1983). The biodegradation of BaP may be inhibited near hazardous waste sites where chemicals toxic to microbes may exist (ATSDR 1990).

Although a large number of studies are available on the persistence of BaP in soil, variation in experimental conditions (such as the matrix, environmental conditions, experimental and analytical techniques) has resulted in poorly defined rates of degradation (Bulman et al. 1985; OMOE 1992). Most studies have reported pseudo-first order degradation. For example, BaP applied to sandy loam soil taken from a site near Toronto, Ontario displayed a half-life of 347 days at an initial concentration of 5 mg·kg⁻¹ and 218 days at an initial concentration of 50 mg·kg⁻¹ (Bulman et al. 1985). Zero order degradation kinetics have also been observed (Bulman et al. 1985). A review of the biodegradation rates of BaP showed rates of transformation ranging from 0.00001 mg·kg⁻¹ day⁻¹ at an initial concentration of 0.01 mg·kg⁻¹ to 161.7 mg·kg⁻¹ day⁻¹ at an initial concentration of 9 100 mg·kg⁻¹ (Sims and Overcash 1983). The role of temperature in biodegradation is not clear. Changes in soil temperature from 10 - 30°C had little effect on the loss of BaP from unacclimated sandy loam soil (Coover and Sims 1987). Estimates of the half-life of BaP in soil vary from 37 - 694 days at low temperatures (<15°C), 30 - 294 days at medium temperatures (15 - 25°C), and 2 - 39 days at high temperatures (>25°C) (Sims and Overcash 1983). Acclimation of soil microbes to BaP greatly affects rates of biodegradation. A study performed under natural conditions showed reductions of BaP of 33 - 71% for acclimated soils and 0% for non-acclimated soils after three months (Sims and Overcash 1983). Partial photolysis of BaP in top soil layers may render the compound more susceptible to microbial attack (Miller et al. 1988). Rates of transformation also depend on the species of microbes, plants and animals present. BaP amended soils showed a consistently higher rate of BaP disappearance in soil units planted with prairie grass than in unvegetated units (Aprill and Sims 1990). BaP biodegradation rates in three soil types (fine sand, medium sand and marsh sediment) ranged from 0.84 - 1.4% loss per week, with rates increasing by a factor of 1.1 - 2.4 following the addition of the polychaete worm Capitella capitata (Gardner et al. 1979 as cited in Sims and Overcash 1983). Little information is available on the biodegradation of BaP in anaerobic soils although preliminary data suggests that the rate is very slow (Barnsley 1975; Bulman et al. 1985).

3.2 Water

Most BaP present in aquatic systems is strongly bound to suspended particles or bed sediments (ATSDR 1990). BaP may also bind to dissolved organic or humic matter within the water column (McCarthy and Jimenez 1985). The majority of PAHs entering rivers, lakes, estuaries and marine environments remain close to their site of entry, suggesting that aquatic systems near human population centres are the primary repositories of aquatic PAHs (Neff 1979; NRC 1983). In general, the cycling of PAHs in aquatic environments is poorly understood (Eisler 1987). Photo-oxidation and biodegradation are considered to be the two most important degradation processes for BaP in the aquatic environment (Suess 1976; ATSDR 1990). A small amount of BaP may be degraded by direct photolysis and chemical oxidation may occur at significant rates when chlorine or ozone exist in sufficient concentrations (ATSDR 1990). BaP in the aquatic environment may also be accumulated in biota (Suess 1976). Volatilization of BaP from aquatic systems is expected to be insignificant in any sizeable body of water due to its low Henry's Law constant ($4.9 \times 10^{-7} \text{ atm}\cdot\text{m}^3/\text{mol}$; ATSDR 1995).

The half-life of BaP in purified water illuminated with light at wavelengths of 313 and 366 nm was

found to be 1.1 and 0.69 hours respectively. Dissolved humic acid was found to reduce the rate of degradation of BaP, while purging oxygen from the water with nitrogen resulted in the almost complete inhibition of photolysis of BaP (Mill et al. 1981). Earlier experimental studies on the photodecomposition of BaP, both in solution and adsorbed onto particles in aqueous suspension, showed that O_2 concentration, temperature, illumination and exposure time increased the rate of degradation, while pH and the ionic strength of the water had no effect (Suess 1976, Bulman et al. 1985). Degradation in natural waters will likely depend on water depth, daily and seasonal changes due to fluctuation of solar radiation, ambient temperature and dissolved oxygen. In addition, the rate of degradation will be higher in the upper layers of the water where illumination, temperature and oxygen concentrations are high. BaP deposited in river, lake and sea sediments are expected to photo-oxidize very slowly, if at all, as a result of the lack of radiation and oxygen (Suess 1976).

Biodegradation is the major fate of sediment-bound BaP (ATSDR 1990), although the rate of sediment biodegradation processes is very slow. The half-life for BaP in sediments has been reported to be 875 days, with half-lives in contaminated streams 10 - 400 times longer (ATSDR 1990). BaP may persist indefinitely in oxygen-poor basins or anoxic sediments (Neff 1979). Marine and lacustrine sediments form the ultimate environmental sinks of BaP (Behymer and Hites 1988).

3.3 Air

BaP in urban and suburban atmospheres is primarily associated with aerosol soot particles less than 3 μ m in diameter (Pierce and Katz 1975; Butler and Crossley 1981). Its dispersion, deposition and residence time in the atmosphere depend upon particle size, type of substrate, meteorological conditions and aerosol dynamics. The major processes affecting transport, distribution and removal of atmospheric BaP are dry and wet deposition and photo-oxidation. Hydrolysis is not considered to be an important fate process for atmospheric BaP as the compound does not contain groups amenable to this reaction (Sims and Overcash 1983).

Dry deposition is the most significant transport process of atmospheric BaP. Approximately 50% of atmospheric BaP reaches the surface via dry deposition, while wet deposition is considered to occur at a much slower rate (ATSDR 1990). In the absence of rainfall, particles less than 1 μ m in diameter in the lower atmosphere have a residence time of four to six weeks, while those with a diameter of 1 - 10 μ m will remain suspended for less than one to several days (Suess 1976). Particles smaller than 10 μ m in diameter are easily inhaled (Grimmer and Pott 1983; Eisler 1987). The atmospheric half-life of BaP as a result of wet deposition alone has been estimated at 1.4 years (Cupitt 1980). Airborne BaP may be transported over relatively long distances from emission sources (Edwards 1983), although deposition rates are much higher near urban centres than in rural areas (Eisler 1987).

Photo-oxidation may be an important processes in the removal of BaP from the atmosphere (Edwards 1983; Eisler 1987). Some of the reaction products of this process may be carcinogenic or mutagenic, although little is known of their persistence (Edwards 1983). BaP strongly absorbs solar radiation at wavelengths above 300 nm (ATSDR 1990). The rate of the photochemical transformation of BaP is complex and dependent upon several variables, including light intensity, concentration of gaseous

oxidants (e.g. singlet oxygen, O_3 , NO_x , SO_x) and the characteristics of the particulates onto which the BaP is adsorbed (Lane and Katz 1977; Valerio et al. 1984). The half-life of atmospheric BaP due to photo-oxidation alone varies from 10 minutes to 72 days, depending on conditions (Valerio et al. 1984). In general, photochemical degradation of BaP will not occur if strong sunlight is not present; therefore, long range transport of BaP is possible under certain conditions (Butler and Crossley 1981).

Considerable controversy exists concerning the rate of degradation of PAHs in air. Some laboratory studies have shown degradation periods as short as a few hours, while other studies indicate little degradation in air and high rates of deposition in soils and sediments (Behymer and Hites 1988). Laboratory studies have shown that the half-life for BaP adsorbed on soot particles in air containing 10 ppm NO_x was 7 days, while little degradation was observed for particulates exposed to ambient laboratory air or air containing 5 ppm SO₂ (Butler and Crossley 1981). Similarly, BaP was found to react readily with NO₂ and SO₃ but not with NO or SO₂ under experimental conditions (Hughes et al. 1980). Ozone was shown to be an effective BaP oxidant, with half-lives in the presence of 0.19 -2.28 ppm ozone ranging from 0.3 - 0.62 hours in the dark and 0.08 - 0.58 hours with illumination (Lane and Katz 1977). An analysis of the effects of substrate type on the degradation rate of BaP showed average half-lives ranging from two hours to more than six days. Darker (black and gray) substrates with high carbon contents tended to stabilize the compound as they absorbed more of the incident radiation (Behymer and Hites 1988). Adsorption onto coal fly ash decreases both the rate and extent of photodecomposition of BaP (Korfmacher et al. 1980a,b). Experimental results confirm that the amount of BaP adsorbed on airborne particulates is negatively correlated to the intensity of sunlight and suggest seasonal variation in atmospheric concentrations (Valerio et al. 1991).

4. BEHAVIOUR AND EFFECTS IN BIOTA

4.1 Soil Microbes

Metabolic Fate and Behaviour

BaP can be metabolized by both fungi (e.g., *Phanerochaete chrysosporium*) and soil bacteria (e.g. *Bacillus megaterium*) (Poglazova et al. 1967; McFarland et al. 1989). Bacteria appear to degrade BaP cometabolically rather than in the substrate utilization mode and the addition of a substrate stimulates BaP degradation (Bossert and Bartha 1986). The metabolism of PAHs by fungi usually proceeds through the monooxygenase pathway and by bacteria through the dioxygenase pathway (Bulman et al. 1985). The biological activity of PAH metabolites is not well known, although the intermediate epoxides formed by the monooxygenase pathway are demonstrated mutagens (Sims and Overcash 1983; Bulman et al. 1985).

4.2 Terrestrial Plants

Metabolic Fate and Behaviour

PAH uptake by plants is largely due to atmospheric deposition onto and absorption through aboveground plant parts (Shabad 1968; Edwards 1983; Sims and Overcash 1983). The degree of PAH contamination of vegetables by this route of exposure depends on the ratio of surface area to mass of the plant as well as on plant location and exposure time (Sims and Overcash 1983). The process of BaP uptake from soils is not well understood but may depend on plant species, BaP concentration, soil type, the presence of solvents and the physicochemical state of the compound (Müller 1976; Edwards 1983; Sim and Overcash 1983; Eisler 1987). BaP absorbed through roots can translocate to other tissues, although rates of uptake are expected to be low as a result of BaP's tendency to bind to organic matter (Edwards 1983; Eisler 1987).

Carrots, radishes and spinach cultivated in two soils (clay-sand/compost and quartz sand) containing 0.041 to 4.208 mg·kg⁻¹ BaP showed significant uptake of the compound. BaP concentrations of 0.0035 mg·kg⁻¹ and 0.020 mg·kg⁻¹ were observed in radish tubers and leaves respectively after seven weeks of growth in clay-sand/compost soil with 2 mg·kg⁻¹ BaP, while radish tubers grown in sand contained a maximum of 0.050 mg BaP·kg⁻¹. Initial soil concentrations of 0.50 mg·kg⁻¹ in each soil type resulted in carrot BaP levels approaching 0.30 mg·kg⁻¹ plant matter in the quartz sand and 0.05 mg·kg⁻¹ plant matter in the compost soil after 17.5 weeks of cultivation. Absorbed BaP was mainly incorporated into the cortical region of the root vegetables (Muller 1976).

Beets, onions and tomatoes grown 50 m south of Highway 401 in Toronto, Ontario received both airborne contaminants and road run-off. The soil of the area was deficient in organic matter, had high clay levels and contained BaP concentrations of 0.108 mg·kg⁻¹ at the soil surface, 0.065 mg·kg⁻¹ at a depth of 5 cm, and 0.087 mg·kg⁻¹ at a depth of 15 cm (Wang and Meresz 1981). Observed levels of BaP in the vegetables included: 0.00736 mg·kg⁻¹ in the peel and 0.00006 mg·kg⁻¹ in the interior of mature onions, 0.00003 mg·kg⁻¹ in the peel and 0.00001 mg·kg⁻¹ in the interior of fresh onions, 0.00021 mg·kg⁻¹ in the peel and 0.00002 mg·kg⁻¹ in the interior of beets, and 0.00001 mg·kg⁻¹ in the peel of tomatoes (Wang and Meresz 1981).

Addition of a BaP/oil solution with a concentration of $3.333 \text{ mg} \cdot \text{kg}^{-1}$ to soil resulted in BaP levels of 0.00013 to 0.0002 mg \cdot \text{kg}^{-1} dry weight in the shoots of rye plants after thirty days, indicating uptake from root to shoot (Montizaan et al. 1989). Wheat cultivated in soil containing 0.170 mg \cdot \text{kg}^{-1} BaP showed no significant uptake of BaP when compared to control plants (Montizaan et al. 1989).

The pathways of BaP metabolism are not well understood although most higher plants can catabolize it (Eisler 1987). Organic acids are the primary products of BaP degradation and are concentrated at the site of assimilation (Sims and Overcash 1983). Other products of metabolism include amino acids, chinons, phenols, diols and CO_2 (Montizaan et al. 1989). Soybean plants were found to metabolize BaP to BaP-9,10-diol and BaP-9-monool (Negishi et al. 1987), while metabolites of BaP degradation in carrots, radishes and spinach included 9,10-dihydro-9,10-dihydroxy-BaP, 4,5-dihydro-4,5-dihydroxy-BaP, 3-hydroxy-BaP, 9-hydroxy-BaP, 3,9-dihydroxy-BaP, BaP-3,6-quinone and BaP- 1,6-quinone (Müller 1976).

Toxicity

A summary of the available toxicological data on the effects of BaP on terrestrial plants is presented in Table 3.

BaP has been shown to both stimulate and inhibit the growth of higher and lower plants (Sims and Overcash 1983; Montizaan et al. 1989). Rye, summer wheat, and green maize grown in soils with BaP concentrations up to 1.2 mg·kg⁻¹ showed no adverse effects (Sims and Overcash 1983). Rye seedlings cultivated in soil containing 3.3 mg BaP·kg⁻¹ showed no adverse effect (Montizaan et al. 1989). Wheat, maize and beans seedlings cultivated in sand with 1.0 - 2.0 mg BaP·kg⁻¹ showed neither stimulation nor inhibition (Montizaan et al. 1989). Soybean seeds soaked for 24 hours in solutions containing 5 - 400 mg·L⁻¹ BaP showed no mutagenic spotting, although abnormal growth and deformed leaf development were observed at the highest concentrations (Fujii and Inoue 1985). Corn seeds soaked for six hours in aqueous solutions containing BaP showed a 14% increase in root growth at concentrations of 0.0005 mg·L⁻¹; this stimulating influence decreased with increasing BaP concentrations until no growth stimulation was observed at 0.02 mg·L⁻¹ (Deubert et al. 1979).

Recent attempts, by Environment Canada (1995), to test the toxicity of BaP on radish (*Raphanus sativa*) and lettuce (*Lactuca sativa*) have produced inconclusive results. In an artificial soil, the highest application rate of 23,800 mg·kg⁻¹ had no effect on radish seedling emergence but inhibited lettuce seedling emergence at the 11,900 mg·kg⁻¹ application rate. Root elongation studies using filter paper in petri dishes showed no effect on either lettuce or radish at the highest application rate of 23,800 mg·kg⁻¹. This test was considered incomplete as attempts to apply higher amounts of BaP in order to produce an effect failed.

4.3 Terrestrial Invertebrates

Metabolic Fate and Behaviour

Very little is known about the uptake, metabolism and elimination of BaP in terrestrial invertebrates (van Straalen and Verweij 1991). Many lower animals do not metabolize PAHs and may therefore accumulate high concentrations of BaP (Sirota and Uthe 1981; Uthe 1991). Salt marsh caterpillars (*Estigmene aerea*) were observed to excrete most ingested BaP as fecal products. Approximately 50% of the 50 μ g BaP fed to two caterpillars was excreted intact, while most of the remainder was degraded by hydroxylation and conjugation to highly polar derivatives (Lu et al. 1977).

Toxicity

Few data have been reported on the toxicity of BaP to terrestrial invertebrates. A summary of the toxicological effects of BaP on terrestrial invertebrates is presented in Table 3.

Isopods (*Porcellio scaber*) fed leaf litter contaminated with 0 - 125 mg·kg⁻¹ BaP showed minimal mortality, all of which were thought to be unrelated to BaP exposure. These results supported previous aquatic toxicity data which suggested a low acute toxicity for BaP (Neff 1979; van Straalen and Verweij 1991). Exposure to the highest level of BaP resulted in a significant increase in the rate of food assimilation and an 84% decrease in the growth efficiency of male animals only. The reasons for these changes were unclear. No other effects related to BaP ingestion were observed (van Straalen and Verweij 1991).

Earthworms (*Eisenia fetida*) exposed to deposits of BaP on filter paper for 48 hours showed an LC_{50} >1000 µg cm⁻² (Roberts and Dorough 1984). Repeated dermal applications of a 0.5% solution of BaP to the earthworm *Lumbricus terrestris* resulted in hyperplasia and incipient tumors both at the application site and at other parts of the body after eight to ten weeks of exposure (Montizaan et al. 1989). Two of 70 apple snails (*Ampullarius australis*) injected with a 1% BaP/oil solution developed papillomas in the area of the operculum (Krieg 1970).

In recent toxicicological studies by Environment Canada (1995), earthworm (*Eisenia fetida*) survival in artificial soil was not affected, even at the highest test application rate of 48 000 mg·kg⁻¹. Such high tolerance in an acute test however may reflect the inappropriateness of such experiments for compounds with documented carcinogenic and mutagenic chronic effects (See section 4.6). The test was considered incomplete as attempts to apply higher amounts of BaP in order to produce an effect failed.

4.4 Mammals and birds

Metabolic Fate and Behaviour

BaP may enter the bodies of terrestrial animals via several routes of exposure, including through the skin and through the ingestion of contaminated food. Wild woodchucks (*Marmota monax*) living near an aluminum electrolysis plant at Jonquière, Quebec and feeding on vegetation containing BaP levels of 7.19 - 35.35 ng·kg⁻¹ dry weight displayed a significantly higher number of BaP-diolepoxide blood protein adducts (a biomarker of environmental contamination by BaP) than animals from the control area. The concentration of albumin adducts was correlated with the level of food contamination (Blondin and Viau 1992). Similarly, detectable concentrations of BaP-hemoglobin adducts were present in various species of small animals (shrews, snapping turtles, rats and muskrats) feeding on vegetation and invertebrates in a floodplain with soil BaP concentrations of 0.070 mg·kg⁻¹ (Shugart et al. 1989). The rate of dermal uptake of BaP in the rat from a soil spiked with 1% crude oil containing 1 mg·kg⁻¹ native BaP was found to be 0.2 ng·cm²·day¹. Only the monolayer of soil in intimate contact with the skin surface was important in the uptake process (Yang et al. 1989).

The metabolism of PAHs in mammalian cells typically proceeds through the monooxygenase pathway (Bulman et al. 1985). BaP is degraded in mammalian systems by the mixed function oxidase system of liver microsomes. This system catalyzes the oxidation of BaP via epoxide intermediates to phenol, diol, and quinone metabolites (Negishi et al. 1987). Genetic injury results from the formation of

adducts by the reactive epoxides and cellular molecules such as DNA, RNA and cellular proteins (Eisler 1987). 7,8-Dihydroxy-9,10-epoxy-7,8,9,10-tetrahydro-BaP is an exceptionally potent carcinogen and is considered to be the ultimate carcinogenic metabolite for BaP (Negishi et al. 1987). Interspecies differences in sensitivity to BaP-induced carcinogenesis are largely due to differences in levels of mixed function oxidase activities which will regulate the rate of transformation of active metabolites to less active products (Neff 1979).

Toxicity

A summary of the available toxicological data on the effects of BaP on mammals, birds and amphibians is presented in Table 4.

The acute oral LD_{50} for BaP in rodents has been estimated at 50 mg·kg⁻¹ (Eisler 1987). Acute and chronic exposure to various carcinogenic PAHs have been observed to result in several types of tissue damage. However, these effects occur at dose levels that would also be expected to induce carcinomas (Eisler 1987). The tissues affected by BaP depend upon the route of exposure and the process of carcinogenesis may occur over a period of many months (Eisler 1987).

Dietary BaP has been demonstrated to lead to leukemia, lung adenoma and stomach tumours in mice, while injection has been found to cause hepatomas (Eisler 1987). Carcinogenic PAHs were found to adversely affect the immune systems of mice. In general, immunosuppression was correlated with the carcinogenicity of the metabolite (Eisler 1987). Skin tumour initiating activities of various BaP metabolites were positively correlated with mutagenic activity in mammalian cell systems (Slaga et al. 1977). Subcutaneous injection of 0.2 mL of Carbowax-400 containing 4 mg of BaP into pregnant mice during the latter half of pregnancy resulted in enhanced tumour formation in offspring (Bulay and Wattenberg 1970). Chicken embryos inoculated with BaP after 3 - 4 and 6 - 7 days of incubation showed sister chromatid exchanges above baseline in a dose-dependent manner. The lowest dose of BaP found to give a significant increase in sister chromatid exchanges was 1.8 μ g per embryo (Bloom 1984). Intraperitoneal injections of 10 to 125 mg BaP·kg⁻¹ body weight into young frogs (*Xenopus laevis*) were found to cause significant alterations in DNA within 60 minutes. Damage decreased to undetectable levels after three hours but reappeared 8 - 12 days after the initial dose (Höhn-Bentz et al. 1983; Zahn et al. 1984). BaP injected into the yolks of preincubated hen eggs at a dose rate of 2 mg·kg⁻¹ resulted in 30% mortality (Brunström 1991).

Many chemicals are known to modify the action of carcinogenic PAHs in laboratory animals, including weakly-carcinogenic and non-carcinogenic PAHs. These modifiers may serve to alter the metabolism and enhance the detoxification of the carcinogen, to scavenge the carcinogens and to prevent access to critical target sites, or to exhibit competitive antagonism (Eisler 1987). Selenium, vitamin E, ascorbic acid, butylated hydroxytoluene, hydrosyanisole, flavones, retenoids and vitamin A have been reported to inhibit PAH-induced tumour development (Eisler 1987).

4.5 Bioaccumulation

BaP is highly lipophilic and can be bioaccumulated to high and potentially hazardous levels in some organisms (Lu et al. 1977). Concentrations of PAHs in vegetation tend to be less than the concentrations in the soil where they grow. Bioaccumulation ratios for plants (defined as the ratio of the concentration in vegetation to the concentration in soil) for BaP range from 0.002 - 0.33 (Edwards 1983). Lower animals, including crustaceans and bivalves, do not metabolize BaP and may bioaccumulate the compound (Varanasi et al. 1985). For example, snails are deficient in microsomal oxidase and are not capable of metabolizing BaP. Snails exposed to 0.0025 ppm BaP for three days bioconcentrated the compound 2177 times (Lu et al. 1977). There has been little documentation on the bioaccumulate in mammalian adipose tissues likely as a result of rapid and extensive metabolisis (Eisler 1987).

4.6 Mutagenicity and Carcinogenicity

Although it is well known that nearly all PAHs are acutely toxic to some degree, concern over the toxicity of PAHs is derived mainly from their highly carcinogenic behaviour. PAHs with three or fewer unsubstituted aromatic rings have not shown carcinogenic behaviour. However, many PAHs with four, five and six unsubstituted aromatic rings are carcinogenic (U.S. EPA 1980; NRCC 1983; Eisler 1987)

PAHs are suspected to be a causative agent of cancer and cancer-like growths in aquatic organisms. Varanasi et al. (1986) reported that the hepatic DNA of starry flounder (*Platichthys stellatus*) and English sole (*Parophrys vetulus*) contained adducts of covalently bound BaP-7,8-diol following benzo(a)pyrene injections. A number of field studies have implicated elevated environmental PAH concentrations as a cause of tumours in fish (Malins et al. 1984; Varanasi et al. 1983)

The carcinogenicity and mutagenicity of BaP have been studied more extensively than the effects of any other PAH. The BaP diol-epoxides have been suggested to be the metabolic intermediates responsible for the induction of tumours and mutations (Varanasi et al. 1986). B(a)P-7,8-diol has been shown to bind to rat DNA and this process is believed to be critical to the promotion of tumours (Varanasi et al. 1986; Slaga et al. 1977). Many studies have suggested that B(a)P-7,8-diol-9,10epoxide is an ultimate carcinogen. It appears that this metabolite can act as a stronger mutagen than benzo(a)pyrene itself and there is no increase in mutagenic activity after metabolic activation (Montizaan et al. 1989; IARC 1983; U.S. EPA 1982).

5. DERIVATION OF ENVIRONMENTAL SOIL QUALITY GUIDELINES

5.1 Introduction

Canadian soil quality guidelines are designed to protect four different land uses: agricultural, residential/parkland, commercial and industrial. The derivation procedures are described in *A Protocol for the Derivation of Environmental and Human Health Soil Quality Guidelines* (CCME 1996).

There were insufficient data available to meet the minimum data requirements described in the Protocol (CCME 1996), for the derivation of soil quality guidelines based on soil contact (SQG_{sc}), soil and food ingestion (SQG_{I}) or the nutrient and energy cycling check for any of the categories of land use. Studies were excluded from use because of one or more of the following reasons:

- soil pH was not recorded;
- soil pH was below 4 (as this is considered outside the normal pH range of most soils in Canada)
- no indication of soil texture was provided;
- inappropriate statistical analysis was used;
- test was not conducted using soil or artificial soil;
- test soil was amended with sewage sludge or a mixture of toxicants;
- test did not use controls.

Therefore, the Provisional Method, described in CCME (1997) and hereafter was used to derive Provisional Canadian Soil Quality Guidelines for BaP. Results are presented in Table 5.

5.2 Provisional Soil Quality Guidelines

Provisional Soil Quality Guidelines Derivation Method

Limitations in the availability of quality soil toxicity data hinder the derivation of soil quality guidelines since the methods described in the Protocol (CCME 1996) are all limited by minimum data requirements. However, litterature searches often yield data which do not meet the requirements of the 1996 Protocol, but still provide some toxicity information. Also, toxicity tests using standard methodologies may produce data that do not meet the regular quality standards defined by toxicologists, due to difficulties in handling and evaluating certain substances such as volatile organic chemicals in the context of a soil contact test for example.

While acknowledging the need for toxicity data of the highest quality, it is the opinion of the Subcommittee that guidelines based on limited toxicity information are still more practical and scientifically defensible than the absence of guidelines or than guidelines that are not risk based. Thus, a provisional method for deriving environmental soil quality guidelines was proposed (CCME 1997). The guidelines derived using this method are called "provisional" to clearly indicate that the underlying data do not meet the requirements of the Protocol (CCME 1996) and that there is an

urgent need for additionnal research. Also, given the increased uncertainty surrounding the provisional soil quality guidelines, such provisional guidelines may not be used to raise an existing criteria or guideline, such as the 1991 interim criteria, but only to lower it if the provisional value indicates that the existing criteria or guideline is not adequately protective.

The guiding principles described in the Protocol (CCME 1996) are to be followed while applying relaxed data requirements in order to allow the derivation of provisional soil quality guidelines. When the data requirements of one of the derivation methods described in the Protocol (CCME 1996) are met, regular soil quality guidelines must be derived. However, when the available data do not meet those requirements, professionnal jugement is used to evaluate the available information and to recommend a provisional soil quality guideline. The derivation of provisional soil quality guidelines is directed by the following landmarks:

- be precautionary: use higher safety factors where degree of uncertainty is high;
- use soil contact data for plants and invertebrates preferentially over mammalian data, equilibrium partitioning data or other data;
- rely on equilibrium partitioning data only to justify additional safety factors and to assist in professional judgement;
- use microbial data as a check mechanism, follow the method described in the protocol (use geometric mean of soil contact number and microbial value where microbial value is lower);
- keep in mind that provisional soil quality guidelines for agricultural and residential/parkland land uses are intended to approximate no appreciable effect levels where those for commercial/industrial land use allow for a low level of effects;
- be consistent with the spirit of the 1996 Protocol.

Derivation of Provisional Soil Quality Guidelines

Because of the strong affinity of BaP for organic matter, food ingestion may contribute a considerable proportion of the benzo(a)pyrene uptake of some soil organisms. It is then desirable to gather data linking food concentrations of BaP to observed effects on soil invertebrates and to derive an equation allowing conversion of food concentrations to soil concentration for the purpose of calculating soil quality guidelines for BaP.

Van Straalen and Verweij (1991) reported a significant response at 125 mg BaP /kg of food for male woodlouse (*Porcellio scaber*) feed poplar leaf litter, the observed effect beeing a decrease in growth efficiency. Furthermore, Jensen and Folker-Hansen (1995) have proposed the following equation to convert food concentrations to soil concentrations. Thus, the soil quality guideline for BaP is calculated as shown below:

$$EC_{soil} = f_{oms} / f_{omf} \cdot EC_{food}$$

where:

- EC_{soil} = concentration of contaminant in soil (mg·kg⁻¹ dry soil)
- f_{oms} = fraction of organic matter in soil : set to 1.70 x f_{ocs} (f_{ocs} = fraction of organic carbon in soil)

f_{omf} = fraction of organic matter in food = 95% (Jensen and Folker-Hansen, 1995) EC_{food} = concentration of contaminant in food = 125 mg BaP /kg (Van Straalen and Verweij, 1991)

If $f_{oc} = 0.3\% = 0.003$ (CCME 1996), then:

$$EC_{col} = 1.70 \times 0.003 / 0.95 \times 125 = 0.67 \text{ mg BaP /kg dry soil}$$

Thus, for agricultural and residential/parkland land uses, the provisionnal environmental soil quality guideline recommended for benzo(a)pyrene is 0.7 mg /kg dry soil.

In the case of commercial and industrial land uses, since the 0.7 mg /kg dry soil value was calculated using the highest concentration tested in the Van Straalen and Verweij (1991) study and that no other similar studies were available, and since it is recognized in the Protocol (CCME 1996) that commercial and industrial land uses do not require the same level of protection as agricultural and residential land uses, an application factor of 0.5 was applied. Thus, for commercial and industrial land uses, the provisionnal environmental soil quality guideline recommended for benzo(a)pyrene is 1.4 mg /kg dry soil.

5.3 Groundwater Check

As a result of its low solubility and high lipophilicity, most of the BaP applied to soil will be very strongly adsorbed to the organic matter of the top soil. According to standard leaching tests, less than 0.2% of the BaP in a contaminated soil was extractable with demineralized water at pH 4 (Pedersen et al. 1993 as cited in Jensen and Folker-Hansen 1995). Hence, it is not likely that BaP will leach to the groundwater in significant amounts, and therefore, the calculation of a soil quality guideline protective of aquatic life through the groundwater check procedure described in CCME 1996 is of little relevance for BaP.

6. DATA GAPS

Generally, very little information on the toxicity of BaP to soil organisms (microbes, invertebrates, and plants) was found. Attempts by Environment Canada (1995) to generate toxicological data for use in the derivation of soil quality guidelines have demonstrated unresolved problems associated with testing BaP. The results of the Environment Canada (1995) study were incomplet. The amount of BaP required to produce an effect in the organisms tested proved to be too high to go into solution for application. These results suggest that acute tests may be inappropriate and that chronic tests may be more informative in evaluating the hazards of BaP to the environment in light of its carcinogenic and mutagenic effects.

Overall, there are insufficient physical, chemical, biological, toxicological and epidemiological data available to reliably evaluate the hazard of BaP to the environment (Suess 1976).

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TABLES

Table 1: Physical and Chemical Properties of Benzo(a)pyrene

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Property	Value				
Synonyms	BaP, BP, 3,4-benzpyrene, 3,4-benzopyrene, benzopyrene, 3,4- benz(a)pyrene, 6,7-benzopyrene, 4,5-benzpyrene, benzo(d,e,f)- chrysene				
Molecular Formula	C ₂₀ H ₁₂				
Molecular weight	252.3 g·mol ⁻¹				
Density	1.351 g/cm ³				
Melting point	179-179.3°C				
Boiling point	310-312°C @ 10 mm Hg				
Vapour pressure	5.6 x 10° mmHg				
Solubility (water)	2.3 x 10 ³ mg·L ⁴				
Henry's Law constant	4.9 x 10 ⁻⁷ atm · m ³ /mol				
Volatilization rate	negligible				
Log K _{ow}	6.06				
Log K _{ee} (organic carbon-water)	5.67* (Mackay et al. 1992)				

Adapted from ATSDR (1995) except where indicated [•] Calculated from the relationship of $K_{oc} = 0.41 \text{ x } K_{ow}$ from Mackay et al. (1992)

Table 2: Existing Criteria and Guidelines for Benzo(a)pyrene in Various Jurisdictions

Jurisdiction	Category	Criterion/ Guideline (mg·kg ⁻¹)	Reference
The Netherlands	Target value	0.025	RIVM 1994
Alberta	Tier 1 criterion for contaminated soil assessment and remediation	1	Alberta Environment Protection 1994
New Jersey	Residential clean-up standard Subsurface clean-up standard	0.66 100	New Jersey Register 1992
British Columbia	Investigation for Agricultural, Residential/Parkland land uses	0.1	B.C. MOE 1989
	Remediation for Agricultural Residential/Parkland Commercial/Industrial	1 1 10	
Québec	A: Background B: Requires site-specific evaluation C: Immediate cleanup	0.1 1 10	MENVIQ 1988
Ontario	Surface soil and groundwater cleanup criteria in a potable groundwater situation Agricultural Residential/Parkland Commercial/Industrial Groundwater (all land uses)	1.2 1.2 1.9 0.01 µg·L ⁻¹	OMOE 1994
	Surface soil and groundwater cleanup in a nonpotable groundwater situation Residential/Parkland Commercial/Industrial Groundwater (all land uses)	1.2 1.9 1.9 µg·L ⁻¹	
	Sub-surface soil cleanup criteria in a potable groundwater situation: Residential/Parkland Commercial/Industrial	1.2 7.2	
	Sub-surface soil cleanup criteria in a nonpotable groundwater situation Residential/Parkland Commercial/Industrial	1.9 7.2	
Canada	Interim remediation criteria Agricultural Residential/Parkland Commercial/Industrial	0.1 1 10	CCME, 1991

Organism	Effect (Exposure period)	Endpoint	Concentration	pH	Test Substrate	Extraction Method	Reference
Invertebrates							
Earthworm (Eisenia fetida)	Mortality (14 days)	NOEC	26 000 mg·kg ⁻¹ (48 000 mg·kg ⁻¹) [*]	4.1	Artificial soil; sand 70%, silt 13%, clay 17%; OM 10.4%.	HNO3, H2O2, HCl w/ AAS	Environment Canada 1995
Earthworm (Eisenia fetida)	Mortality (2 days)	LC ₃₀	>1000 µg·cm ⁻²	NR	Filter paper contact test.	NR	Roberts and Dorough 1984
Plants							·····
Soybean (Glycine max)	Mutagenic spotting (24 hours) Abnormal growth and deformed leaf	NOEC LOEC	400 mg·L ^{·1} 400 mg·L ^{·1}	NR	Seeds soaked in BaP	NR	Fujii and Inoue 1985
Lettuce (Lactuca sativa)	Root elongation (3 days)	NOEC	17 500 mg·L ⁻¹ (23 800 mg·L ⁻¹)	NR	Filter paper	HNO3, H2O2, HCI w/ AAS	Environment Canada 1995
Radish (Raphanus sativa)	Root elongation (3 days)	NOEC	17 500 mg·L ⁻¹ (23 800 mg·L ⁻¹)	NR	Filter paper	HNO3, H2O2, HCI w/ AAS	Environment Canada 1995
Lettuce (Lactuca sativa)	Seedling emergence (5 days)	NOEC LOEC (40% reduction)	4400 mg kg ⁻¹ (6000 mg kg ⁻¹) 8800 mg kg ⁻¹ (11 900 mg kg ⁻¹)	4.1	Artificial soil; sand 70%, silt 13%, clay 17%; OM 10.4%	HNO ₃ , H ₂ O ₂ , HCl w/ AAS	Environment Canada 1995
Radish (Raphanus sativa)	Seedling emergence (3 days)	NOEC	17 500 mg·kg ⁻¹ (23 800 mg·kg ⁻¹)	4.1	Artificial soil; sand 70%, silt 13%, clay 17%; OM 10.4%	HNO3, H2O2, HCl w/ AAS	Environment Canada 1995

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' Values in parenthesis indicate nominal concentrations.

NR = Not reported.

Table 4. Available Vertebrate Toxicity Studies for Benzo(a)pyrene

Organism	Effect (Exposure period)	Endpoint	Concentration (mg·kg ⁻¹ bw)	Reference
Mouse	Frequency of adenomas in progeny (3 subcutaneous doses during gestation)	LOEC	4	Bulay and Wattenberg 1970
Chick embryos	Mortality (Single dose in egg yolk)	LOEC (30% mortality)	2	Brunström 1991
Frog	Frequency of DNA alteration (Single subcutaneous dose)	LOEC	10	Höhn-Bentz et al. 1983

Table 5. Summary of Environmental Soil Quality Guidelines for Benzo(a)pyrene

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Guideline	Land Use				
	Agricultural (mg ·kg ')	Residential/ Parkland (mg ·kg ⁻¹)	Commercial/ Industrial (mg -kg ⁻¹)		
TEC or ECL (a)	Insufficient data.	Insufficient data.	Insufficient data.		
Nutrient and Energy Cycling Check	Insufficient data.	Insufficient data.	Insufficient data.		
SQC _{sc}	Insufficient data.	Insufficient data.	Insufficient data.		
SQC ₁	Insufficient data.	Insufficient data.	Insufficient data.		
Provisional SQC _E	0.7	0.7	1.4		
Interim Criteria (CCME 1991)	0.1	1	10		

(a) As per CCME (1996) protocol, the SQG_{sc} for Agricultural and Residential/Parkland land uses correspond to the 25th percentile of the effects and no effects data distribution when using the Weight of Evidence method, while the SQG_{sc} for Commercial/Industrial land use corresponds to the 25th percentile of the effects only data distribution.



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CANADIAN SOIL QUALITY GUIDELINES FOR BENZO(A)PYRENE: ENVIRONMENTAL