ENVIRONMENTAL BEHAVIOUR AND FATE OF DICHLORODIPHENYLTRICHLOROETHANE (DDT) RESIDUES IN A TERRESTRIAL ARCTIC ECOSYSTEM

by

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ABSTRACT

The environmental behaviour and fate of DDT (dichlorodiphenyltrichloroethane) and its related compounds DDE and DDD were examined in a terrestrial arctic ecosystem. Samples of soil, sediment, willow (*Salix* sp.), grass (*Elymus* sp.), and arctic ground squirrel (*Spermophilus parryi*) were collected at an abandoned Long Range Aid to Navigation (LORAN) station located at Kittigazuit, Northwest Territories (69°16'55.71"N, 133°54'31.80"W). Since the study site received applications of DDT pesticide between 1948 to 1950, it provided a unique opportunity to examine the legacy of localized DDT use in a terrestrial arctic environment.

Despite the passage of time, soil concentrations have remained high (maximum ΣDDT 210000 $ng \cdot g^{-1}$), and the composition of ΣDDT compounds in soil still resembles the original pesticide formulation (59% *p,p'*-DDT). In soils, appreciable loss and degradation of DDT was less pronounced than compared to temperate and tropical environments. The cold climate had greatly influenced the behaviour and fate of DDT. Due to the lack of degradation, contaminated soils continued to act as sources for DDT contamination in the ecosystem.

Contaminant concentrations in willows and grasses demonstrated the availability of soil contaminants. The concentrations and compositions of ΣDDT in both willows and grasses were consistent with plants growing in contaminated soils in temperate and tropical climates; DDT accumulation was low and there was little change in composition. Calculated bioaccumulation factors were low for both genera, and differences between genera were due to different lipid concentrations. Contamination of plants indicated a route of food chain exposure between soil and animals.

The concentration and composition of ΣDDT in arctic ground squirrels livers were clearly the result of contamination at the study site. Liver concentrations at contaminated areas (maximum ΣDDT 4300 $ng \cdot g^{-1}$) declined to background levels (maximum ΣDDT 4.5 $ng \cdot g^{-1}$) with increasing distance from contaminated areas. Estimated contaminant exposures were below no-observed effect levels, but a significant relationship between liver size and ΣDDT concentration was found. It was suggested that the ecology of arctic ground squirrels could enhance the toxicological effect of ingested contaminants. In conclusion, the environmental behaviour and fate of DDT in a terrestrial arctic ecosystem is governed by the cold climate.

CO-AUTHORSHIP

Research to determine the extent of DDT contamination at the study site was conducted by the Environmental Sciences Group (ESG) in partnership with the Inuvialuit Lands Administration (ILA). Funding was provided by the Contaminated Sites Office of the Department of Indian Affairs and Northern Development (DIAND). ESG and ILA were jointly responsible for sample collection. Axys Analytical Services Ltd. analysed sediments, plants and animal tissues. Queen's University Analytical Services Unit, as well as the author, conducted analysis of soils.

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TABLE OF CONTENTS

ABSTRACT .... **i**

CO-AUTHORSHIP .... **iii**

ACKNOWLEDGEMENTS .... **iv**

I. GENERAL INTRODUCTION **1**

II. LITERATURE REVIEW **4**

A. OVERVIEW 4
1. Toxicity *6*
2. Chemical Properties *7*
3. Degradation Pathways *8*
a) Microbial 9
b) Mammalian 10
B. ENVIRONMENTAL BEHAVIOUR AND FATE 11
a) Soils and Sediments 11
b) Terrestrial Plants 18
c) Mammals 19
C. STUDY SITE 24

III. MATERIALS AND METHODS **28**

A. SAMPLE COLLECTION 28
1. Soils *28*
2. Sediments *30*
3. Plants *30*
4. Mammals *31*
B. SAMPLE ANALYSES 31
1. Soils *31*
2. Sediments *33*
3. Plants *34*
4. Mammals *35*
C. DATA ANALYSES 36

IV. RESULTS **41**

A. SOILS AND SEDIMENTS 41
B. PLANTS 49
C. MAMMALS 53

V. DISCUSSION **63**

A. SOILS AND SEDIMENTS 63
B. PLANTS 70
C. MAMMALS 71
D. CONCLUSIONS 83

LITERATURE CITED **86**

APPENDIX A: MAPS OF SAMPLE LOCATIONS **94**

APPENDIX B: DATA **106**

VITA **113**

LIST OF TABLES

Table II-1: Chemical properties governing the environmental behaviour of ΣDDT compounds (modified from EC, 1998a). 8

Table III-1: Input parameters for calculation of TDI for *S. parryi*. 39

Table IV-1: Concentrations of ΣDDT in surface soils collected from background locations, the station and the camp (n=2, 97 and 19, respectively). The applicable federal soil criterion is provided for comparison. 42

Table IV-2: Concentrations of ΣDDT in *Elymus* sp. and *Salix* sp. samples collected from background locations, the station and the camp (n=2, 13 and 3 for *Elymus* sp. and n=3, 16 and 3 for *Salix* sp., respectively). 49

Table IV-3: Compound-specific bioaccumulation factors for *Elymus* sp. and *Salix* sp. (n=16 and 19, respectively). Factors are expressed on both a plant dry weight to soil dry weight and a plant wet weight to soil dry weight basis 53

Table IV-4: Comparison of physical characteristics between sexes of *S. parryi* collected from all areas. Values are expressed as means (± standard deviation). 54

Table IV-5: Concentrations of ΣDDT in *S. parrvi* collected from a background location, the station, the camp and the access road (n=2, 11, 3, and 7, respectively). 55

Table IV-6: *p,p'*-DDT BAFs for arctic ground squirrels. BAFs were calculated using wet weight tissue concentrations (not lipid corrected) and dry weight plant and soil concentrations. Liver concentrations of animals at the station and camp (n=7 and 3, respectively) were divided by the range of plant or soil concentrations at either the station or camp. 59

Table IV-7: Monte Carlo simulation results for calculation of TDI of DDT through the ingestion of either *Salix* sp. or *Elymus* sp. at the station or camp. Probability of exceeding NOEL for each scenario is also provided for each scenario 60

Table IV-8: Expected *p,p'*-DDT and ΣDDT liver burdens based on calculated TDI values for different diets at the station and camp. 61

Table IV-9: Expected *p,p'*-DDT and ΣDDT liver burdens based on soil-leaf-mammal pathway BAF of 0.067[1] 62

Table V-1: Initial and final ΣDDT composition in various soil studies. Studies are ranked according to increasing latitude. 68

Table V-2: Published DDT BAFs for plants 71

Table V-3: Published concentrations and compositions of ΣDDT compounds in the livers of various mammals. Samples collected from areas with and without use of DDT pesticides. Time elapsed between final application and sample collection is indicated. 75

Table V-4: Published DDT BAFs from plant tissue-animal tissue and soil-animal tissue food chain for rodents. 77

LIST OF FIGURES AND ILLUSTRATIONS

Figure II-1: Chemical structures of the six ΣDDT compounds: a) *p,p'*-DDT, b) *o,p'*-DDT, c) *p,p'*-DDE, d) *o,p'*-DDE, e) *p,p'*-DDD and f) *o,p'*-DDD. 4

Figure II-2: Degradation relationship between DDT, DDE and DDD. 9

Figure II-3: Metabolites of *p,p'*-DDT and the postulated route of metabolism in the rat. Compounds indicated by an asterisk have also been identified in humans (from Smith, 1991). 11

Figure II-4: Location of study site at Kittigazuit, Northwest Territories (69°16'55.71"N, 133°54'31.80"W). 25

Figure III-1: Sample locations for sediments, plants, and ground squirrels. 29

Figure IV-1: Spatial extent of ΣDDT contamination at the station. Distribution based on data for surface soils. Contaminant concentrations are in $ng{\cdot}g^{-1}$ 43

Figure IV-2: Spatial extent of ΣDDT contamination at the camp. Distribution based on data for surface soils. Contaminant concentrations are in $ng{\cdot}g^{-1}$ 44

Figure IV-3: Relative ΣDDT composition of surface soil samples collected from both the camp and station (n=116). Compositions are expressed as the percent contribution of a specific compound to the total detectable concentration of ΣDDT. The composition of TG-DDT indicated by stars. The top and bottom of each box indicate the interquartile range and the horizontal line represents the median. Vertical lines indicate data within 1.5 interquartile ranges from either box edge. Asterisks represent data between 1.5-3 interquartile ranges and circles represent data beyond 3 interquartile ranges. 45

Figure IV-4: Relative ΣDDT composition of sediment samples collected from water bodies at both the camp and station (n=4). Compositions are expressed as the percent contribution of a specific compound to the total detectable concentration of ΣDDT. 46

Figure IV-5: Principal components loading (top) and score (bottom) plots of the relative ΣDDT compositions of surface soil samples from both the camp and station areas (n=116). Values in parentheses indicate percent of total variance explained. For the first component, the regression relationship was described by: first principal component = 1.871 – 0.638($\log_{10}$ ΣDDT $ng{\cdot}g^{-1}$ soil dry weight) (linear regression, $r^2 = 0.332$, $p < 0.001$, n = 114). For the second component, the regression relationship was described by: second principal component = – 1.166 + 1.597($\log_{10}$ percent organic content) (linear regression, $r^2 = 0.348$, p = 0.002, n = 24). 48

Figure IV-6: Relative ΣDDT composition of *Elymus* sp. (left) and *Salix* sp. (right) samples collected from both the camp and station (n=16 and 19, respectively). Compositions are expressed as the percent contribution of the specific compound to the total detectable concentration of ΣDDT. 50

Figure IV-7: Principal components loading (top) and score (bottom) plots of the relative ΣDDT compositions of *Elymus* sp. and *Salix* sp. samples from the station and camp areas (n 35). Values in parentheses indicate percent of total variance explained. 52

Figure IV-8: Relative ΣDDT composition of *S. parryi* collected from all areas (n=21). Compositions are expressed as the percent contribution of a specific compound to the total detectable concentration of ΣDDT. 56

Figure IV-9: Principal components loading (top) and score (bottom) plots of the relative ΣDDT compositions of *S. parryi* samples from all areas (n=21). Values in parentheses indicate percent of total variance explained. For the first principal component: principal component 1 = -0.079 - 0.617($\log_{10}$ ΣDDT $ng \cdot g^{-1}$ lipid) (linear regression, $r^2=0.481$, $p<0.001$, n=19). For principal component 2: principal component 2 = -1.374 + 0.044(liver mass g wet weight) (linear regression, $r^2=0.395$, $p<0.005$, n=19). 58

Figure IV-10: Relationship between LSI, and ΣDDT for both sexes of *S. parryi* from all locations. LSI was found to increase with increasing concentrations of ΣDDT. For ΣDDT: LSI = 0.041 + 0.007 ($\log_{10}$ ΣDDT $ng \cdot g^{-1}$) (linear regression, $r^2 = 0.450$, $p = 0.002$, n = 18). 62

I. GENERAL INTRODUCTION

DDT (dichlorodiphenyltrichloroethane) is a chlorinated organic pesticide that is known to be a toxic and persistent pollutant (ASTDR, 1994; EC, 1998a). In 1945, DDT pesticides became available to the Canadian public and were used primarily for agricultural purposes (EC, 1998a). As early as the 1950s, it was noted that DDT and its related compounds, DDE (dichlorodiphphenyldichloroethylene) and DDD (dichlorodiphenyldichloroethane), were able to bioaccumulate and biomagnify in the environment (Lawless, 1977). Recently, Canada classified DDT as a Track 1 substance under the federal Toxic Substances Management Policy because it is persistent, bioaccumulative, as well as anthropogenic and is therefore targeted for virtual elimination from the environment (EC, 1997).

At present, DDT is distributed in the atmosphere, water, sediments, and soils world-wide (EC, 1998a). Biota in each of these environmental compartments are affected, with toxic effects in reptiles, mammals, and birds being well documented (EC, 1998a). Most studies on the behaviour and fate of DDT have focused on temperate or tropical environments where there has been extensive localized use of DDT pesticides, with little attention given to northern areas. The long-range transport of DDT, DDE, and DDD to northern environments, however, has received much attention (Braune et al., 1999).

The Arctic is particularly susceptible to the impacts of persistent organic pollutants, such as DDT, given the ecology of its organisms and its climate. Arctic biota are extremely dependent on lipid reserves, which are also the preferred sites for DDT accumulation. The persistence of organochlorines in the polar environment is increased

due to low temperatures, limited biological activity and the relatively low incident sunlight (Falconer et al., 1995). Consequently, DDT contamination in northern environments is not directly comparable to contamination in other regions. Many studies have focused on DDT transported to the Arctic from temperate and tropical environments, whereas information on the environmental behaviour and fate of DDT at a local point-source in the Arctic is rare. In fact, in the most recent soil quality guidelines for DDT residues, Environment Canada identified a data gap and stated that, "data on soil residual levels [of DDT] in Canada, particularly in northern Canada, are scarce if not non-existent" (EC, 1998a).

The present study was conducted to address this data gap; it examined the environmental behaviour and fate of DDT in a terrestrial Arctic ecosystem. The research was conducted at an abandoned Long Range Aid to Navigation (LORAN) station located on the Mackenzie River delta at Kittigazuit, Northwest Territories. The LORAN station was occupied from June 1948 to March 1950, during which time DDT pesticide was liberally sprayed at the LORAN station (Hart & Cockney, 1999). The study was designed to investigate DDT, DDE and DDD residues at the site, and to track their movement though a short terrestrial food chain.

The specific objectives of this thesis were:

1. To examine environmental behaviour and fate of DDT pesticide in soils, sediments, plants, and animals in a terrestrial arctic environment.
2. To model the potential exposure of arctic ground squirrels (*Spermophilus parryi*) to DDT contamination through the ingestion of soil, willows (*Salix* sp.), and grasses (*Elymus* sp.).

Following this introduction, a review of current literature is presented in Chapter II, followed by an explanation of materials and methods in Chapter III. Combined results for both objectives are presented in Chapter IV, and discussed in Chapter V.

II. LITERATURE REVIEW

A. Overview

DDT (dichlorodiphenyltrichloroethane) generally refers to a chlorinated organic pesticide comprised of two phenyl groups, as well as pesticide formulations containing DDT as the primary active ingredient. DDT consists of *p,p'*-DDT [1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane] and its isomer *o,p'*-DDT. *p,p'*-DDE [1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene] and its isomer *o,p'*-DDE, as well as *p,p'*-DDD [1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane] and its isomer *o,p'*-DDD are closely related compounds of DDT. The term ΣDDT refers to all six of these compounds (Figure II-1).

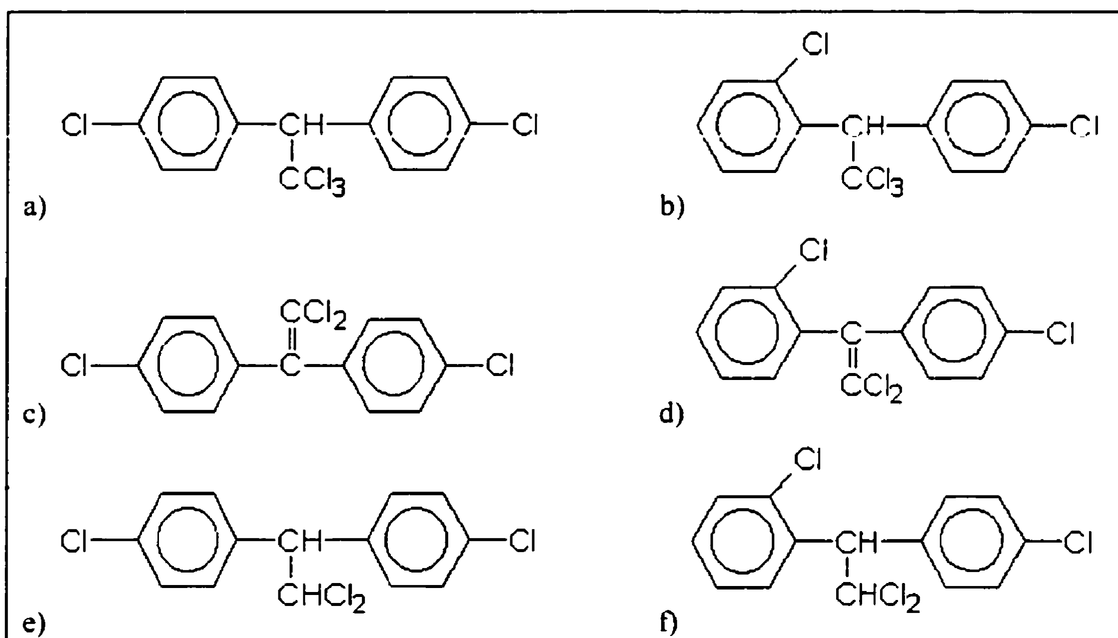

Figure II-1: Chemical structures of the six ΣDDT compounds: a) *p,p'*-DDT, b) *o,p'*-DDT, c) *p,p'*-DDE, d) *o,p'*-DDE, e) *p,p'*-DDD and f) *o,p'*-DDD.

DDT was first synthesized in Germany in 1874. World War II marked the beginning of large-scale manufacture and application of DDT pesticide, first as a body

louse powder and later as an effective agent to deal with insect-borne diseases (Lawless, 1977). When the war ended, global use of DDT increased, primarily due to is application as an agricultural pesticide, but also in human health programs such as the Global Malaria Eradication Campaign (Trigg & Kondrachine, 1998). During 1945, DDT pesticides became available to the Canadian public (EC, 1998a). Within a decade, the deleterious nature of DDT, DDE and DDD in the environment became evident (Lawless, 1977).

Although DDT was never manufactured in Canada, it was imported from the United States until 1985 (EC, 1998a). In Canada, formal restrictions pertaining to DDT products were first enacted under the Pest Control Products Act and Regulations. Under this legislation, the registration of all uses of DDT pesticides was suspended in 1985; however, existing stocks could be used until December 1990, after which DDT pesticides were banned (EC, 1998a). Recently, the Canadian Toxic Substances Management Policy classified DDT as a Track 1 substance requiring its virtual elimination from the environment, specifically because it is persistent, bioaccumulative, and anthropogenic (EC, 1997). Despite severe restrictions or bans in most industrial nations, DDT pesticides continued to be produced, exported, and used in many tropical countries (EC, 1998a).

The continued use of DDT pesticides is of global concern. ΣDDT compounds have a demonstrated potential for long-range transport. According to the global distillation hypothesis, sequential volatilisation and condensation of organic pollutants results in their transport towards the poles. This process has been cited as an important source of contaminants in Canada's northern environment (Mackay & Wania, 1995). In fact, the persistence, bioaccumulation, toxicity, and long-range transport of DDT has

made it a candidate for international control initiatives through organisations such as the United Nations (EC, 1998a).

1. Toxicity

As a result of their high lipid solubility and low water solubility, ΣDDT compounds are retained in fatty tissue where they remain immobile and relatively harmless until the fat is metabolized (WHO, 1989). Accumulation rates vary with species, duration and concentration of exposure, and with environmental conditions (WHO, 1989). The life histories of organisms can exacerbate the toxic effects of ΣDDT residues. Species exhibiting marked seasonal cycles in fat content are most vulnerable to toxic effects. The main effect on such mammals is to increase the mortality of adults during periods of stress (WHO, 1989). Natural periods of stress during an animal's life are events such as migration or hibernation.

A variety of toxic effects following exposure to DDT pesticides are documented. Adverse effects on reproduction, growth, and immunocompetence have been observed in both mammalian and avian species exposed to ΣDDT compounds (ASTDR, 1994). Long-term dietary exposure can result in mutagenic and carcinogenic effects in various species (ASTDR, 1994). ΣDDT residues can reduce longevity, alter cellular metabolism, disrupt neural activity, and alter liver function (EC, 1998a).

There is no doubt that DDT and a number of other chlorinated hydrocarbon insecticides cause marked changes in the livers of various rodents and that these changes progress to tumour formation in some species (Smith, 1991). DDT is an inducer of mixed function oxidase (MFO) enzymes of the liver (ASTDR, 1994). These enzymes are components of the biological defence of living organisms against chemical stresses in the

environment, and function by adding oxygen to organic compounds, rendering them more soluble and more easily excreted by the body (Fox, 1993). Recent reviews of the chronic toxicity of orally-administered DDT in mammals agree on a no-observed effects level (NOEL) of approximately 0.37 mg DDT·kg^{-1}bw·day^{-1} for CF-1 mice, which are the most sensitive herbivorous species to DDT exposure (Smith, 1991; EC, 1998a). This NOEL is in accordance with other estimates of the threshold for induction of various enzymes in the rat (Smith, 1991).

2. *Chemical Properties*

Commercially produced DDT pesticides contain a mixture referred to as technical grade-DDT (TG-DDT). TG-DDT contains 77.1% *p,p'*-DDT, 14.9% *o,p'*-DDT, 4.0% *p,p'*-DDE, 0.1% *o,p'*-DDE, 0.3% *p,p'*-DDD, 0.1% *o,p'*-DDD, and 3.5% unidentified compounds (WHO, 1989). The primary active ingredients of DDT pesticides are always *p,p'*-DDT and *o,p'*-DDT; DDE and DDD are considered by-products of the manufacturing process (EC, 1998a). At 25°C TG-DDT is a waxy solid, but for applications it was formulated into solutions in xylene or petroleum distillates, emulsifiable concentrates, water-wettable powders, granules, aerosols, smoke candles, chargers for vaporizers, and lotions (WHO, 1989).

Three main chemical properties of ΣDDT compounds are responsible for their environmental behaviour: i) vapour pressure, ii) water solubility, and iii) octanol-water partition coefficient (log K_{ow}). Similar to other persistent and bioaccumulative organochlorines, ΣDDT compounds have relatively low vapour pressures and water solubilities, but high log K_{ow} values (Table II-1) (EC, 1998a). The K_{ow} value predicts the partitioning behaviour of a hydrophobic organic substance between being dissolved in

water and being sorbed by solid organic matter associated with the water (vanLoon & Duffy, 2000). Also, K_{ow} values are related to the tendency of compounds to become associated with lipids in biological tissues because octanol can dissolve lipophilic compounds (Kenaga, 1980; vanLoon & Duffy, 2000). This tendency for compounds to become associated with biological tissue is bioaccumulation and can be expressed using bioaccumulation factors (BAFs). BAFs refer to the uptake of a compound from all sources of exposure and are based on field measurements (CCME, 1997). BAFs are calculated as follows:

$$BAF = \frac{\text{concentration of chemical in organism in the field}}{\text{concentration of chemical in the diet/soil}}$$

Table II-1: Chemical properties governing the environmental behaviour of ΣDDT compounds (modified from EC, 1998a).

Property	**Compound**					
	***p,p'*-DDT**	***o,p'*-DDT**	***p,p'*-DDE**	***o,p'*-DDE**	***p,p'*-DDD**	***o,p'*-DDD**
Vapour Pressure[1] (mPa)	0.02	0.02	1	0.8	0.1	0.2
Water Solubility[1] ($\mu g \cdot L^{-1}$)	3	3	40	100	50	100
log K_{ow}	6.0	6.0	5.7	5.8	5.5	6.1

1. At 20°C.

3. Degradation Pathways

The active ingredients of TG-DDT, *p,p'*-DDT and *o,p'*-DDT, can be transformed into their corresponding DDE or DDD isomers. The pathways may proceed from DDT to DDE, and then to DDD, or from DDT to DDD directly (Corona-Cruz et al., 1999). Production of DDE and DDD can occur through both biotic and abiotic mechanisms. DDE is formed from DDT through photochemical reactions in the presence of sunlight, and through dehydrochlorination in bacteria and animals (Aislabie et al., 1997).

Similarly, DDD is formed from DDT through reductive dechlorination, either directly by microbially mediated processes, or as the result of chemical reactions mediated by biomolecules (Aislabie et al., 1997) (Figure II-2). Though the products of DDT degradation are known, their biochemical pathways have not been completely established, even after 40 years of study (Hayes, 1991).

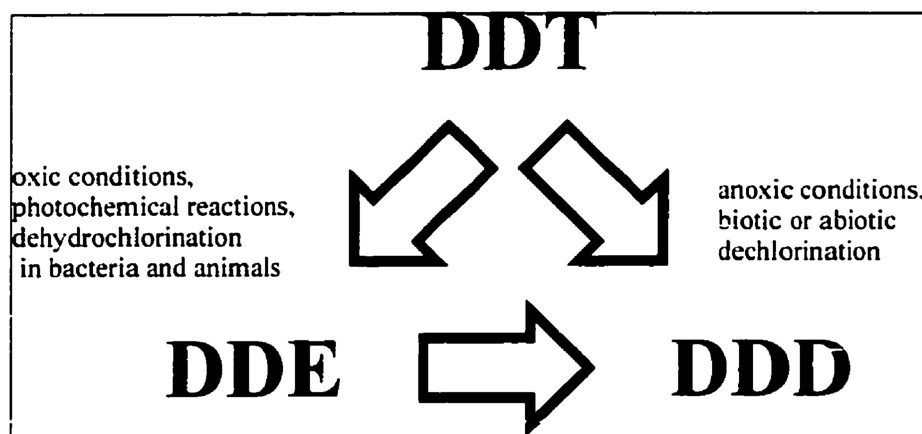

Figure II-2: Degradation relationship between DDT, DDE and DDD.

a) Microbial

A number of microbes can degrade DDT to DDD in soils and sediments (Aislabie et al., 1997), but the microbial processes leading to the formation of DDE are less well defined. Under aerobic conditions, the predominant reaction is dehydrochlorination of DDT to yield DDE (Aislabie et al., 1997). Under anaerobic conditions, transformation of DDT to DDD by reductive dechlorination is considered to be the dominant reaction (Aislabie et al., 1997). There are no definitive reports of the aerobic biotransformation of DDT to DDD (Aislabie et al., 1997).

The transformation of DDT to DDD occurs readily in soils under certain conditions, and the process may be attributed directly to microbial activity, either bacterial or fungal, or indirectly due to the generation of anaerobic conditions (Aislabie et al., 1997). In the presence of an alternative carbon source, numerous bacteria and fungi are capable of converting DDT to DDD (Aislabie et al., 1997).

b) Mammalian

The biochemical pathway for DDT metabolism in rodent systems is relatively consistent across taxa. As with other lipophilic xenobiotics, DDT can be metabolized by the microsomal cytochrome P-450 system (MFO enzymes) to hydroxyl derivatives (Smith, 1991). In mammals the major urinary metabolite of DDT is 2,2-bis(*p*-chlorophenyl) acetic acid (DDA), which can be produced from either DDE or DDD intermediates (Figure II-3) (Smith, 1991). In rats, DDE is slowly converted in the liver to 1-chloro-2,2-bis(*p*-chlorophenyl)ethene (DDMU) and then to DDA by way of 2,2-bis(*p*-chlorophenyl)ethane (DDNU) (ASTDR, 1994). DDD is rapidly detoxified by way of DDMU to 1-chloro-2,2-bis(*p*-chlorophenyl)ethane (DDMS) and then to DDNU; DDNU is then further metabolized, primarily in the kidney, to 2,2-bis(*p*-chlorophenyl)ethanol (DDOH) then to 2,2-bis(*p*-chlorophenyl)ethanol (DDHCO) or to DDA.

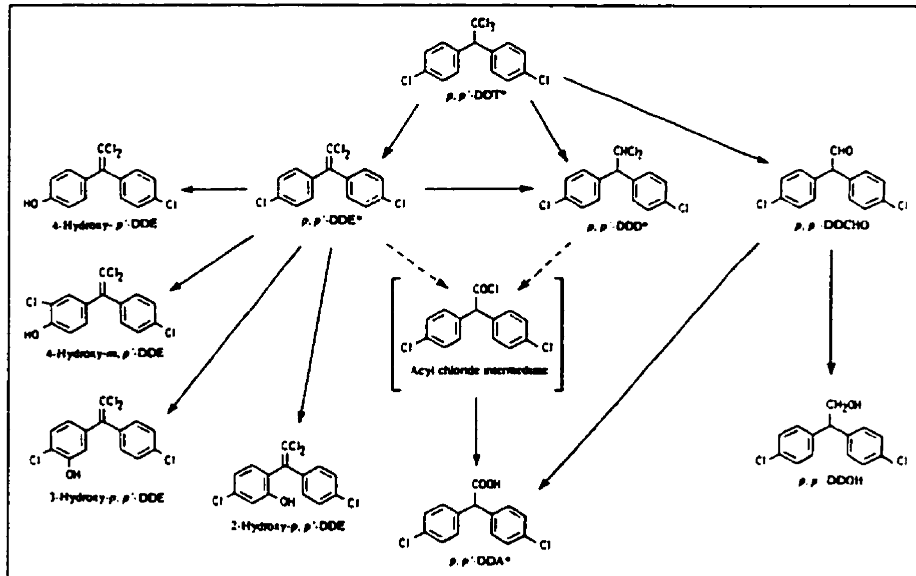

Figure II-3: Metabolites of *p,p'*-DDT and the postulated route of metabolism in the rat. Compounds indicated by an asterisk have also been identified in humans (from Smith, 1991).

B. Environmental Behaviour and Fate

a) Soils and Sediments

The environmental behaviour and fate of ΣDDT compounds in soil is well documented. ΣDDT residues are resistant to breakdown and are readily adsorbed to sediments and soils, which can act both as sinks and as long-term sources of exposure (WHO, 1989). In these compartments, DDT is slowly converted to DDE and DDD, as well as to a number of other products (Boul et al., 1994). Upon discharge into the soil, the behaviour and fate of ΣDDT compounds depends on three general factors: properties of the specific ΣDDT compound, properties of the soil, and the temperature (Nicholls, 1991). Interactions among these three factors drastically alter the behaviour and fate of ΣDDT compounds in different soils and environments. For any pesticide, potential fates

can include wind drift, runoff, leaching, root uptake, adsorption, chemical degradation, photodecomposition, microbial degradation, and volatilisation (Aislabie & Lloyd-Jones, 1995). The relative importance of these fates can change with time. For example, loss via wind drift refers to the loss of pesticide during the application process such that the pesticide fails to reach its target area. The contribution of wind drift as a potential fate is less relevant when discussing the long-term behaviour of pesticide residues, except in situations where soil particles might be redistributed aerially.

The physiochemical properties of ΣDDT compounds can also alter the relative importance of certain fates. The contribution of runoff, leaching, and root uptake are often dismissed when explaining the loss of ΣDDT compounds from soils. Loss of ΣDDT compounds due to runoff or leaching is negligible considering the low solubility of ΣDDT compounds in water. The loss of ΣDDT residues in runoff is primarily due to transport of particulates to which these compounds are bound, and not by dissolution in water. The low mobility of ΣDDT compounds in soil is supported by long-term field studies. Using irrigated plots dating from 1949, Boul et al. (1994) failed to find downward movement of residues in the soil profile as would be expected if significant leaching occurred; but instead a trend of decreasing residue levels with depth was evident. In general, the uptake of ΣDDT compounds by plants is low, and unless the plants are physically removed from the site, the compounds will re-enter the soil system when the plant decomposes. Loss of ΣDDT compounds via runoff, leaching, and root uptake, are of less importance than loss due to adsorption, chemical degradation, photodecomposition, microbial degradation, and volatilisation.

Over time, an apparent loss of ΣDDT compounds occurs as pesticide molecules become adsorbed to soil particles. During these extended periods of time, the ΣDDT compounds undergo extensive adsorption to soil particles (EC, 1998a). The adsorption of DDT by various types of soils has been investigated and found to be lowest in sandy loam, intermediate in clay soil, and greatest in organic soils. Adsorption of DDT was closely related to organic matter, and humic matter was specifically identified as the major source of adsorptive capacity for DDT (WHO, 1989). Less is known about the binding of DDT to clay (Boul, 1995). As a result of the ΣDDT compounds being bound strongly to soil, they are not easily displaced from the site of application and appreciable amounts may remain in soil for extended periods of time (ASTDR, 1994). The binding of DDT to soil is a matter of some ecological and toxicological importance. Once bound to soil, it appears that DDT residues are detoxified and lose their activity, but residues can still accumulate in animals many years after application (Aislabie et al., 1997).

Even in oxic conditions, localized anoxic environments can occur. Soil becomes anoxic when it is too compacted for oxygen to permeate, or when there is decomposable organic matter present anaerobic conditions can develop (Nicholls, 1991). As a result, DDT degradation to both DDE and DDD occurs under field conditions. Forest soils sprayed with DDT from 1958 to 1967 were sampled for persistence of residues at subsequent intervals and most recently in 1993 at which time the biotransformation compounds DDE and DDD each comprised approximately a third of the total residues (Dimond & Owen, 1996).

Environmental conditions must be suitable such that degradation can proceed. For most pesticides, influential factors can include: pH temperature and salinity, available

water, oxygen tension and redox potential, nutrient availability, presence of alternative carbon substrates, light quality and intensity, binding surfaces, and alternative electron acceptors (Aislabie & Lloyd, 1995). In the case of DDT degradation, redox potential and temperature are the most influential factors.

Volatilisation of DDT, DDE, and DDD is known to account for a considerable loss of these compounds from soil surfaces. Volatile loss is most pronounced immediately following DDT application, and with certain land practices. Despite their low vapour pressure and solubility, chemicals such as DDT are subject to evaporative loss (Sunito et al., 1988). Of all ΣDDT compounds, *p,p'*-DDE has a volatility tenfold greater than *p,p'*-DDD, and fifty-fold greater than *p,p'*-DDT. A field test of the rate of disappearance of DDT from soil near Lake Nakuru, Kenya, found that DDT sublimed directly without prior degradation to DDE (Sleicher & Hopcraft, 1984). In India, high soil temperature, intense sunlight and humidity were identified as the major factors responsible for dissipation by volatilisation (Samuel & Pillai, 1989). In tropical climates, volatilisation is the predominant fate for DDT. In sandy loam soil, loss through volatilisation has been found to increase five-fold when the temperature increased from 15 to 45°C (Samuel & Pillai, 1989). Loss due to volatilisation differs among climates on the basis of such temperature differences. Correction for climatic temperature has been applied to a northern environment to explain the partitioning of ΣDDT among air, water, sediments, and soils in Lake Baikal, Russia. Following correction from standard conditions to an average temperature of 2°C, volatile loss for *p,p'*-DDT reduced by a factor of five and *p,p'*-DDE reduced by a factor of ten (Iwata et al., 1995).

A review of the literature indicates discrepancies regarding degradation rates of ΣDDT compounds in soil. Half-life measurements under field conditions for aerobic degradation of DDT in soil range from two years (Lichenstein & Shulz, 1959) to more than 15 years (Keller, 1970; Stewart & Chisholm, 1971). In flooded soil or under anaerobic conditions, biodegradation is faster, with half-lives estimated from 16-100 days (Castro & Yoshida, 1971). When volatilisation was the predominant means of DDT loss, half-lives were as low as 100 days (Sleicher & Hopcraft, 1984).

Difficulties in accurately assessing DDT half-life results from the multitude of natural degradation mechanisms, multiphase structure of the natural environment, and the interplay of degradation and partitioning via multiple phase-transfer and mixing dynamics (Muller-Herold, 1996). Studies applying an exponential model to describe DDT disappearance from soil have clearly pointed out the need to use half-life values rationally with a clear understanding of their limitations (Sleicher & Hopcraft, 1984; Samuel et al., 1988; Samuel & Pillai, 1991). As noted by Sleicher and Hopcraft (1984), the idealized case of sublimation from a single smooth, flat area with a uniform deposit of DDT would give a constant mass-transfer rate and a linear decay of concentration as long as the area of the deposit remained constant. In actual fact, the degradation of pesticides rarely is linear, nor does it follow first-order kinetics in most cases (Scheunert, 1993). Though the exponential law is not a good model of DDT disappearance under all circumstances, it often provides a useful measure of disappearance rate provided that the time span is not too long and that it is understood that the half-life is a function of many other variables (Sleicher & Hopcraft, 1984).

Soil characteristics such as moisture and organic carbon content can influence the fate of ΣDDT compounds. Laboratory experiments have shown that flooding enhances the loss of DDT from soil due to a number of mechanisms including the creation of anaerobic micro-environments for microbes to degrade DDT via DDD, abiotic reductive dechlorination, and the binding of DDT residues to soil particles (Aislabie et al., 1997). Alteration of mean moisture levels also dramatically affects residue levels in field studies. Experimental trials with irrigated soils resulted in ΣDDT levels 40% of non-irrigated soils, when mean moisture in the top 10 cm of soil only differed by 13.5% during the summer (Boul et al., 1994). Researchers argued that increased soil moisture could result in the creation of anaerobic microenvironments for micro-organisms able to degrade DDT via DDD, or abiotic reductive dechlorination of DDT to DDD could occur in such environments.

In soils, pesticide molecules may bind tightly to soil organic matter, thereby reducing biological availability. Adsorption is significantly correlated with soil organic carbon, and the octanol-water partition coefficient is the preferred physiochemical parameter to predict soil adsorption (Scheunert, 1993). Pesticide adsorption can enhance microbial degradation in soil. The concentration of pesticide applied to the soils might be toxic to microbes, but once the pesticide becomes bound to soil an apparent reduction in toxicity allows biodegradation to proceed (Aislabie & Lloyd-Jones, 1995).

A number of studies report a rapid rate of reduction of DDT to DDD in soil under reducing conditions when a readily available energy source such as alfalfa, barley straw, or glucose is present (Aislabie et al., 1997). For example, fertilization with superphosphate resulted in lower soil levels of *p,p'*-DDT and ΣDDT residues compared

to unfertilized controls (Boul et al., 1994). Although the enhanced degradation of DDT following addition of organic matter to soils indicates the significance of co-metabolic transformations, this effect could also arise because of microbial production and release of porphyrins and/or the generation of anaerobic conditions resulting from enhanced microbial growth (Aislabie et al., 1997).

In an Arctic climate, the persistence of organochlorines is increased due to low temperatures, limited biological activity and the relatively low incidence of sunlight (Falconer et al., 1995). A single study documents the behaviour and fate of DDT pesticide use in an Arctic environment. A 41 km^2 area surrounding Fort Churchill underwent experimental DDT applications at 0.28 $kg{\cdot}ha^{-1}$ for at least four years between 1947 and 1954, followed by subsequent aerial spraying for mosquito control at 0.25 $kg{\cdot}ha^{-1}$ twice annually from 1955-1963 (Brown & Brown, 1970). Approximately three years later, ΣDDT concentration was 88 $ng{\cdot}g^{-1}$ and consisted of 67% DDT, 19% DDE, and 13% DDD.

In temperate and tropical aquatic sediments, behaviour and fate of DDT are less variable than in soils. DDT becomes associated with settling particles and is removed to the benthic region where it may be recycled or incorporated into sediments (Swackhamer & Eisenreich, 1991). Once DDT is removed from the water column through sedimentation processes, it is vulnerable to microbial degradation (Chernyak et al., 1995). Once incorporated into benthic sediments, degradation from DDT to DDD predominantly occurs.

Lakes receive atmospheric and terrestrial input from their entire catchment basins and can effectively concentrate contaminants in sediments at detectable levels. ΣDDT

compounds have been detected throughout lake sediments in the Arctic, but decline with latitude. Muir et al. (1995) collected sediment cores from eight remote lakes in Canada, along a mid-continental transect from 49°N to 82°N. ΣDDT levels in surface sediments declined significantly with latitude from 6.3 and 9.7 $ng \cdot g^{-1}$ at 49°N, to 0.1 $ng \cdot g^{-1}$ at 81°N (Muir et al., 1995). ΣDDT concentrations observed by Mudroch et al. (1992) in Great Slave Lake sediments were 0.25-1.2 $ng \cdot g^{-1}$ and a major fraction (48-60%) of ΣDDT in surface sediments consisted of the dechlorination product, *p,p'*-DDD.

b) Terrestrial Plants

Terrestrial plants can act as significant pathways for DDT exposure to receptors in the ecosystem. Uptake into plants is the first step towards the bioaccumulation of DDT in the terrestrial food web (Trapp, 1993). Three main pathways for chemical movement from soils to plants exist: i) root uptake into conduction channels and subsequent translocation, ii) uptake from vapour in the surrounding air, and iii) uptake by external contamination of shoots by soil and dust, followed by retention in the cuticle or penetration through it (Bell & Failey, 1991).

Despite being strongly bound to soil, DDT, DDE, and DDD can be bioavailable to plants (ASTDR, 1994). Verma and Pillai (1991) reported that grain, maize, and rice plants can accumulate soil-bound residues of DDT. The majority of residues were found in roots of plants, and the lowest concentration of DDT residues was found in shoots, indicating low translocation of DDT. Ware et al. (1970) found that the epidermal layer of alfalfa roots contained five times the amount of DDT in whole roots and six times that found in the cortex, which suggests that DDT and/or its degradation products become bound to the root epidermis and thus cannot move inward.

Soil characteristics can influence the behaviour and fate of ΣDDT compounds in plants. Fuhremann and Lichtenstein (1980) applied ^{14}C-labelled *p,p'*-DDT to loam or sandy soil and grew oat plants on treated soils for 13 days. Very little DDT (0.2% of the total DDT applied) and none of its metabolites, were detected in oat roots grown in loam. Uptake was greater (4.6%) in roots of oats grown on sand, but uptake of labelled carbon into plant tops, from both soils, was below detection limit. The low uptake of DDT by plants is consistent across taxa. Experimentally-derived BAFs for DDT residues in plants are generally below 1.00 and often below 0.50 (Jongbloed et al., 1996).

c) Mammals

Factors that influence the bioaccumulation of DDT, and subsequent BAF values, in mammals include species sex, age, duration of exposure, parental body burden, metabolic capacity to degrade DDT, and lipid content (Borrell & Aguilar 1987; WHO, 1989; Tanabe et al., 1994). While DDT is distributed throughout mammalian tissues, it accumulates preferentially in tissues with high lipid contents (EC, 1998a). Of the tissues commonly sampled, muscle typically has the lowest concentrations of DDT (Muir et al., 1992; Ronald et al., 1984).

In terrestrial ecosystems, exposure can occur via soil contact, inhalation of vapours, ingestion of contaminated water, soil ingestion, and ingestion of contaminated food. Given the chemical properties of ΣDDT compounds, exposure via contaminated water and vapours are typically omitted. DDT is poorly absorbed by the skin from solutions, and the absorption of solid material is so poor that it is difficult or impossible to measure either the uptake of DDT or its effect (Smith, 1991). Therefore, exposure via dermal contact and subsequent absorption is not considered to be a significant exposure

pathway. The most significant pathways are soil ingestion and ingestion of contaminated food.

For animals, inadvertent soil ingestion is an everyday occurrence related to ingestion of soiled plants, coat and muzzle licking, and inhalation of dust (Sheppard, 1998). The significance of this pathway depends on the amount of soil ingested, the concentration on the ingested soil, and the bioavailability of the contaminant on the ingested soil (Sheppard, 1998). The amount of soil adhering to plants can range from as low as 0.03-4 g dry soil kg^{-1} dry plant for grains and tall plants, to as high as 450 g dry soil kg^{-1} dry plant for short annual plants (Sheppard, 1998).

Field-collected data show that the highest levels of DDT are generally observed in terrestrial predators (EC, 1998a). After treating a field ecosystem at a dose rate of 0.92 $kg{\cdot}ha^{-1}$, Forsyth and Peterle (1973) measured DDT residues in various tissues of two species of shrews (*Blarina brevicauda* and *Sorex cinereus*) during a three year study. The highest residues (135 $mg{\cdot}kg^{-1}$) occurred in fat, compared with 10, 10, and 4 $mg{\cdot}kg^{-1}$ in liver, muscle, and brain, respectively.

Environmental monitoring following the single application of DDT to a forest to control spruce budworm at a rate of 0.89 $kg{\cdot}ha^{-1}$, found residues of DDT and metabolites in mammals for over nine years (Dimond & Sherburne, 1969). Herbivorous mice, voles and hares contained less DDT than carnivorous mink and insectivorous shrews. In herbivores, tissue residues approached pre-treatment levels after six to seven years; whereas residues remained elevated in shrews and mink after nine years likely because of their higher position in the food chain. In these species, the authors calculated that it would take at least 15 years for residues to reach background levels. Contaminated soil

was identified as the long-term source of high residue levels in mammals since there was little retention of ΣDDT compounds in vegetation (Dimond & Sherburne, 1969).

Among terrestrial mammals, the highest levels of DDT have been observed in species that include fish and other aquatic animals in their diet (EC, 1998a). Fishers (*Martes pennanti*), martens (*Martes americana*) and mink (*Mustela vison*) collected from southern Ontario from 1972-1974 had muscle tissue DDE concentrations of 61 $ng{\cdot}g^{-1}$, 18 $ng{\cdot}g^{-1}$, and 54 $ng{\cdot}g^{-1}$, respectively (Frank et al., 1979). Lower concentrations of DDT (<10 $ng{\cdot}g^{-1}$) were observed in the muscle tissues of foxes (*Vulpes fulva*), racoons (*Procyon lotor*), and skunks (*Mephitis mephitis*) taken from same location.

The predominance of *p,p'*-DDE in the liver is considered to be indicative of past exposure, and has been observed in martens (*Martes americana*) and fishers (*Martes pennanti*) from the Algonquin region of south-central Ontario (Steeves et al., 1991). The Algonquin region is a forested area of 43,000 km^2 on the Precambrian shield, and has no major industrial or agricultural development. DDT was used in the 1950s and 1960s to control biting insects around tourist establishments (Steeves et al., 1991). Sampling in 1981 indicated that ΣDDT composition was dominated by DDE. The livers of 125 martens contained 84% DDE, 11% DDT and 5% DDD; similarly the livers of 123 fishers contained 71% DDE, 20% DDT, and 9% DDD (Steeves et al., 1991).

The regional concentration of ΣDDT in the Arctic has been studied in caribou (*Rangifer tarandus*). Caribou are well suited to determine background levels for contaminants in the Arctic since their foraging ranges represent large areas. Caribou from five locations across the Northwest Territories representing herds from Bathurst, Quamanirjuaq, Southampton Island, Cape Dorset and Lake Harbour contained levels of

ΣDDT in fat ranging from 0.46 ng·g^{-1} in Arviat caribou to 2.58 ng·g^{-1} in Cape Dorset caribou (Elkin & Bethke, 1995). Musk-ox, and caribou livers from Banks Island and Prince of Wales Island, respectively, did not contain DDT-related compounds at a detection limit of 0.05 ng·g^{-1} (Thomas et al., 1992). The five caribou herds examined by Elkin and Bethke (1995) contained predominantly *p,p'*-DDE. Similarly, ΣDDT compositions in otter and mink collected from forested areas in northeastern Alberta contained only DDE, and DDD was not detectable in either tissues at the analytical detection limit of 0.5 ng·g^{-1} (Somers et al., 1987).

On the basis of food chain transfer of persistent organic pollutants, such as ΣDDT, obligate carnivores are expected to accumulate higher background concentrations. As carnivores are at the terminus of their respective food chains, semi-aquatic mammals such as marten, and mink might be expected to accumulate pesticides even at low environmental concentrations. Poole et al. (1995), examined organochlorine contaminants in harvested mink and marten along the Mackenzie River drainage basin in western Northwest Territories. Despite the absence of historical DDT use at the sample locations, liver tissue levels were elevated. Concentrations of ΣDDT in the livers of mink ranged from 1.24 ng·g^{-1} to 13.66 ng·g^{-1} and were 2.41 ng·g^{-1} in marten (Poole et al., 1995). Residues in liver tissue were the result of long-range transport of ΣDDT compounds to northern Canada.

The study by Brown and Brown (1970) of DDT used near Churchill, Manitoba is the only study of a known point source of ΣDDT in the Arctic. Samples were collected from two areas (treated and controlled) three years after the last treatment. From the treated area, livers of collared lemmings (*Dicrostonyx groenlandicus*) contained 680

$ng \cdot g^{-1}$ ΣDDT, comprised of 3.7% DDT, 70.9% DDD and 25.4% DDE. Liver tissue of red squirrels (*Tamiasciurus hudsonicus*) contained 537 $ng \cdot g^{-1}$ ΣDDT, composed of 3.0% DDT, 54.4% DDD, and 42.6% DDE. In the untreated area, the livers of collared lemmings contained 52 $ng \cdot g^{-1}$ ΣDDT, comprised of 17.3% DDT, 48.1% DDD and 34.6% DDE. Liver tissue of red squirrels contained 57 $ng \cdot g^{-1}$ ΣDDT, composed of 19.3% DDT, 49.1% DDD, and 31.6% DDE.

At known point-sources of contamination, small mammals with limited home ranges are well suited for monitoring the environmental behaviour and fate of contaminants. This study focuses on arctic ground squirrels (*Spermophilus parryi*) to examine the behaviour and fate of DDT pesticide residues in a terrestrial arctic ecosystem. The ecology of ground squirrels makes them well suited to studying an organochlorine compound such as DDT.

A physiological adaptation of many arctic animals to harsh winters is the deposition and utilisation of fat reserves (Batzli, 1981). As a consequence, such animals are susceptible both to the accumulation of persistent organic compounds and to the potential for enhanced toxicity caused by mobilisation of fat reserves which may be laden with lipophilic organic compounds (Geyer et al., 1990; Shore & Douben, 1994). This situation holds for grounds squirrels. Ground squirrels rely heavily on fat reserves as an energy source during their eight to ten months of hibernation (Yukon Department of Renewable Resources, 2000). The well-studied foraging patterns of ground squirrels allow for the modelling of contaminant exposure from their home-range.

Arctic ground squirrels live in colonies along river banks and lake basin rims, with home-ranges between 1.5 to 4.3 ha (Batzli & Sobaski, 1980). Ground squirrels are

opportunistic herbivores, with 40 species of herbaceous diccotyledons comprising 25% to 75% of the diet. Most foraging occurs within 50 m of burrow sites (Batzli & Sobaski, 1980). Feeding during the summer is intense, and by late August, the proportion of body weight as fat can exceed 50% (Batzli & Sobaski, 1980; Kiell & Millar, 1980). The ecology of arctic ground squirrels makes them ideal bioindicators of DDT contamination.

C. Study Site

This thesis focuses on the environmental behaviour and fate of DDT pesticide at the abandoned Long Range Aid to Navigation (LORAN) station located at Kittigazuit in the Northwest Territories (69°16'55.71"N, 133°54'31.80"W) on Kuururyuaq Creek, in the Mackenzie River delta (Figure II-4). Based on nearby climate data from Tuktoyaktuk (69°27'N, 133°00'W), the annual mean temperature and precipitation are low, -10.5 °C and 142.1 mm, respectively (EC, 1998b).

In 1946, construction and testing of a low-frequency LORAN system in the Canadian Arctic was initiated as a joint venture between the Royal Canadian Air Force (RCAF) and the United States Air Force (USAF) (Hart & Cockney, 1999). The Kittigazuit LORAN station was operational between June 1948 and March 1950 (Hart & Cockney, 1999). At the height of operations, station infrastructure consisted of 33 buildings including a pump house, garage, warehouses, boiler house, three interconnected buildings, as well as a 192 m transmitting tower, water line, and a gravel access road between the station and the river. Current infrastructure at the site consists of the shell of one building, the collapsed remains of the LORAN transmission tower, a number of foundations, and building pads.

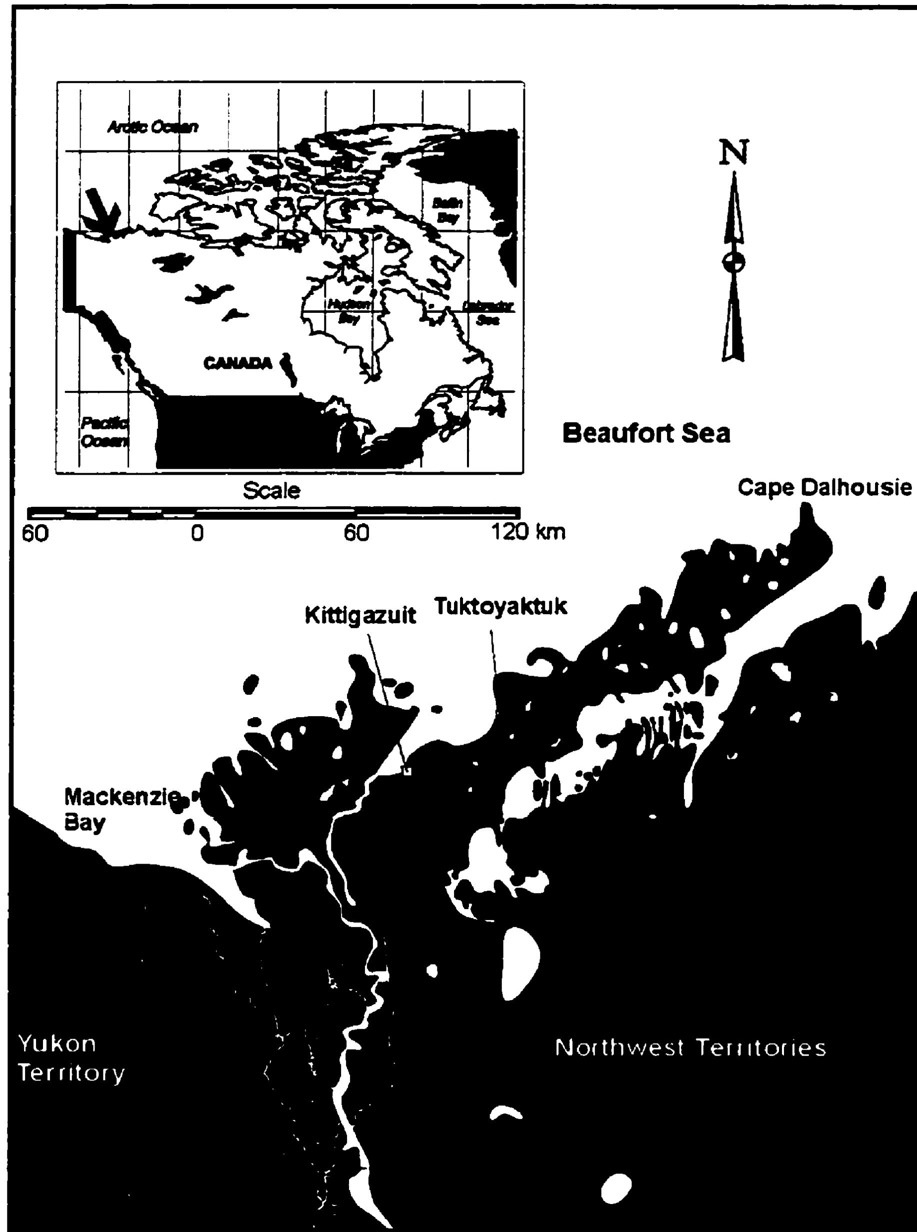

Figure II-4: Location of study site at Kittigazuit, Northwest Territories (69°16'55.71"N, 133°54'31.80"W).

To control mosquito populations, DDT was liberally sprayed in the vicinity of the station (Hart & Cockney, 1999). The daily use of DDT was captured on a station diary entry from July 5th, 1948, which reads as follows:

> Whole of the camp area is sprayed at least once per day to control pests. Inside of windowsills have strips of cloth soaked in DDT solution tacked on to aid in pest control. Solution added to cloth daily to maintain moisture content. Report made to NWAC that "D Dust" not satisfactory due to being non-soluble enough to pass through sprays at this unit (Hart & Cockney, 1999).

In addition to the station, a small camp which was the initial base of operations for the reindeer herding industry in the western Canadian Arctic, was settled in 1935, approximately 1 km west of the LORAN station (Hart & Cockney, 1999). The camp was constructed in 1930 to house Norwegian Saami herders following a five-year reindeer drive from Alaska. During the operation of the LORAN station, Aboriginal workers and their families occupied these cabins at the camp (Hart & Cockney, 1999). During the same period, Aboriginal workers maintained a tent camp immediately south of the cabins.

At present, the reindeer herding camp consists of three log cabins, a plank shack, and a building foundation. No records indicating the spraying of DDT in the vicinity of the cabins were found. Both the station and camp are now under the jurisdiction of the Department of Indian Affairs and Northern Development (DIAND) Contaminated Sites Office, Northern Affairs Program (ESG 1999).

In 1998, the Environmental Sciences Group (ESG) was commissioned by DIAND to conduct an environmental site assessment and delineation of the station and camp (ESG, 1999). Samples were screened for a variety of inorganic and organic contaminants. Analytical results of soil samples indicated the presence of ΣDDT residues in surface soils. Also, results for plants indicated that ΣDDT residues were entering the food chain (ESG, 1999).

Given this initial finding, DIAND contracted a preliminary Ecological Risk Assessment (ERA) based on the data from the ESG investigation (Inuvialuit Environmental, 1999). Calculations were based on a limited data set of soil concentrations, and estimated plant concentrations, which generated predicted tissue concentrations in receptors. Based on the available data, the ERA concluded that ΣDDT residues posed a low risk to arctic ground squirrels. Also, the ERA recommended the measurement of tissue concentrations in plants and animals, and the measurement of bioconcentration factors for DDT in plants and animals (Inuvialuit Environmental, 1999). To that end, ESG returned to the site and collected samples, which formed the basis of this thesis (ESG, 2000).

III. MATERIALS AND METHODS

A. Sample Collection

Soil and plant samples were collected in August 1998. In July 1999, animal and sediment samples were collected, as well as additional soils and plants. During the second visit, sampling focused on areas that had been identified as contaminated during the first visit. On-site samples were obtained among infrastructure, and from visibly disturbed areas at the station and the camp, and off-site samples were taken at a minimum distance of 500 m from the station (Figure III-1) (Appendix A)[1].

Sample locations for soils, plants, and animals were assigned unique numeric codes, and were recorded using an Ashtech® Super CA 12 Precision GPS receiver with post-processing. Locations were recorded in Universal Transverse Mercator (UTM) co-ordinates and measured to the nearest centimetre (Appendix B)[2].

1. Soils

Soil samples were collected as either surface or depth samples. Surface samples represented a composite of the top 10 cm of the soil horizon, whereas depth samples were collected at specific intervals to a maximum depth of 60 cm. Soil samples were collected using a sterile plastic (on-site samples), or metal (off-site samples) scoop, and placed into 125 mL amber glass jars. Excessive organic matter, such as roots, twigs, mosses and leaves, as well as pebbles were excluded from soil samples. Prior to sampling, metal scoops were oven-dried at 300°C for eight hours and wrapped in oven-dried aluminium foil to ensure scoops were free of organic material. A manual soil auger was used to

[1] Appendix A contains detailed maps of sample locations for soils, plants, and animals.
[2] Appendix B contains specific UTM co-ordinates as well as analytical data.

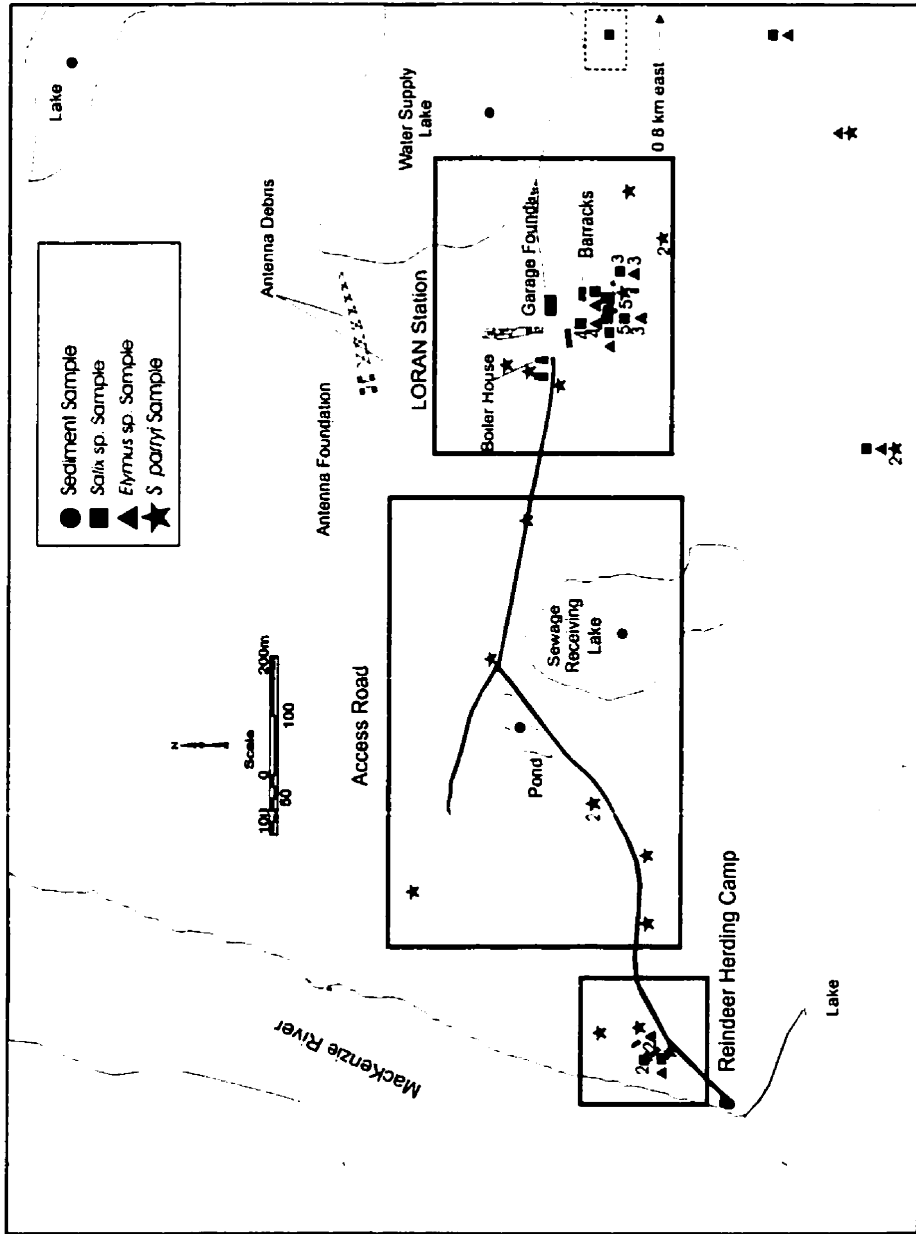

Figure III-1: Sample locations for sediments, plants, and ground squirrels.

collect depth samples. Cross contamination between depth samples was minimized by removal of soil adhered to the auger between samples, and rejecting the initial samples of soil from the new location.

A deterministic sampling strategy was employed at both the station and the camp. Sampling was focused around building locations, within drainage pathways, and in areas of historical activity, such as debris storage areas. Samples were catalogued, and stored in coolers containing ice-packs.

2. Sediments

Sediment samples were collected from the three lakes surrounding the station, as well as a pond adjacent to the road between the station and camp. A Ponar® grab was used to collect samples of the top 5 cm of sediment. Sediments were transferred from the Ponar® grab using a sterile plastic scoop, and placed in 125 mL amber glass jars. Samples were catalogued, and stored in coolers at approximately 4°C.

3. Plants

Two genera of plants were collected: *Salix* sp. (i.e. willow) and *Elymus* sp. (i.e. grass). Plant samples were collected as whole specimens, inclusive of root and shoot portions. These samples represented the dominant vegetation in the area, as well as potential forage for animals. Samples of *Salix* sp. and *Elymus* sp. were collected from areas on-site and off-site. Where possible, samples of both genera were collected from the same location.

Plants were removed using hand tools, and temporarily stored in Ziploc® bags. Within 24 hours of collection, plants were washed thoroughly, and rinsed using potable water. Specimens were dried using absorbent towels, wrapped in sterile aluminium foil, and sealed in clean Ziploc® bags. Samples were catalogued, and frozen.

4. Mammals

Arctic ground squirrels (*Spermophilus parryi*) were collected using three techniques. In visibly populated areas, large Victor® rat traps, and Havahart® live animal traps were baited with raisins. Traps were placed at the entrance to burrows, among bushes, and along runs between burrows. Animals trapped alive were killed by cervical dislocation. A .22-calibre rifle was used to shoot specimens at ranges of less than 50 m. Length, body mass, liver mass, and sex were recorded for each specimen regardless of the collection method. Whole livers were excised using dissection tools, which were cleaned with hexane between specimens. Livers were wrapped in sterile aluminium foil, and sealed in clean Ziploc® bags. Samples were catalogued, and frozen.

B. Sample Analyses

1. Soils

In total, 155 soil samples were analysed. The majority of soil samples were analysed by the Queen's University Analytical Services Unit in Kingston, Ontario. Wet soil was homogenized, and a separate sub-sample was dried to determine moisture content. Samples were run in batches of about 20, with a blank, control, and sample duplicate placed in each batch. Blanks were prepared with 40 g of anhydrous sodium sulphate and 20 g of Ottawa sand, and then spiked with 100 μL of decachlorobiphenyl. Controls were prepared as above, but were also spiked with 100 μL of a prepared pesticide spike. For each sample, 15 g of homogenized, wet soil was added to 40 g of anhydrous sodium sulphate, 20 g of Ottawa sand, and mixed until free flowing.

Samples were extracted by soxhlet for 4-6 hours, at 4-6 cycles per hour using 250 mL of methylene chloride. The extract was concentrated by roto-evaporation to 1 mL,

after which 5 mL of hexane was added, and again evaporated to 1 mL. This solvent exchange was repeated twice more resulting in 1 mL of hexane solvent, and the resulting solution was applied to a Florisil® column. The column was thoroughly rinsed with hexane and the eluant diluted to 10 mL. A set of 3 surrogate standards using 100 µL of decachlorobiphenyl, brought to a final volume of 10 mL, was prepared. Fractions were analysed by gas chromatography/mass spectrometry (GC/MS).

GC/MS analysis used an HP 5890 Series II Plus gas chromatograph equipped with an HP 5972 Mass selective detector, a PTE-5 fused silica capillary column (30 m, 0.25 mm ID x 0.25 µm film thickness), and HPChem station software. GC/MS carrier gas was helium at a flow-rate of 1 $mL \cdot min^{-1}$. One characteristic ion plus the retention times for each target compound and surrogate were used for identification. Results obtained for *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, and *o,p'*-DDD were expressed as nanograms of pesticide per gram dry weight of soil ($ng \cdot g^{-1}$) (Appendix B). The analytical detection limit for this method was 20 $ng \cdot g^{-1}$ for each ΣDDT compound. Sample results at analytical detection limits were set to half of the detection limit. Precision was monitored with field and analytical duplicates. For field sampling, 10% of soil samples were collected in an identical time and fashion from a single sampling location to serve as a duplicate. For analytical techniques, 5% of samples analysed were duplicates within each batch.

An estimation of organic matter in soils was determined by loss on ignition (ASTM, 1998). A random sub-sample of 25 soil samples were selected for analysis. Soil samples were allowed to air dry overnight and about 2 g of dry soil was oven dried at 105°C for 24 hours. Dried samples were weighed and placed in a Fisher Scientific®

muffle furnace 650 series for 1.5 hours at 420°C. Weights of resulting ash were recorded, and the difference between the two weights was used to determine the percent organic matter.

2. Sediments

In total, four sediment and two soil samples were analysed using high-resolution GC/MS by Axys Analytical Services Limited in Sidney, British Columbia. Wet soil was homogenized, and a sub-sample was dried for moisture determination. For each sample, 15 g of homogenized, wet soil was spiked with aliquots of a surrogate standard solution containing ^{13}C-labled surrogates of *p,p'*-DDE, *p,p'*-DDT, and PCB 101, and shaken with 1:1 dichloromethane:methanol solution on a shaker table for 20 minutes. The extract was filtered and the particulates were re-extracted with dichloromethane. The extraction process was repeated three times. Following removal of the methanol phase, the dichloromethane extract was dried over anhydrous sodium sulphate for 15-20 minutes. The extract was transferred to a Kuderna-Danish flask and rinsed with dichloromethane. The extract was concentrated and transferred to a centrifuge tube with hexane rinses and concentrated prior to column chromatography.

The extract was applied to a calibrated Florisil column. The column was eluted with hexane followed by 15:85 dichloromethane:hexane. The eluates were collected together, the combined fraction was concentrated to a small volume, and an aliquot of recovery standard solution containing ^{13}C-labelled PCB 153 was added prior to GC/MS analysis.

GC/MS analysis was performed using a VG 70SE mass spectrometer equipped with a HP 5890 GC and a CTC autosampler. Chromatographic separation was achieved with a

DB-5 column (60 m, 0.25 mm ID x 0.1 μm film). Concentrations of target analytes were calculated by comparing the area of the quantitation ion to that of the corresponding ^{13}C-labelled standard, and correcting for response factors. Concentrations of analytes were corrected based on the percent recovery of surrogate standards. Results were obtained for all six compounds of ΣDDT, and expressed as nanograms of pesticide per gram dry weight of soil or sediment ($ng \cdot g^{-1}$) (Appendix B). The analytical detection limit for this method was sample-specific and determined by surrogate recovery efficiency. Reported concentrations were corrected using surrogate recovery efficiency. Sample results at analytical detection limits were set to half of the sample-specific detection limit. The precision of analytical techniques was monitored with sample duplicates within an analysis batch.

3. *Plants*

In total, 40 plants were analysed in this study, specifically 18 samples of *Elymus* sp. and 22 samples of *Salix* sp. All plant samples were analysed by Axys Analytical Services Limited in Sidney, British Columbia. Wet tissue was homogenized, and a sub-sample dried for moisture determination. For each sample, 15 g of homogenized, wet tissue, and anhydrous sodium sulphate was ground with a glass mortar and pestle to a free-flowing powder, and spiked with a surrogate standard solution containing containing ^{13}C-labled *p,p'*-DDE, *p,p'*-DDT, and PCB 101. The mixture was transferred to a glass chromatographic column containing dichloromethane and eluted with additional solvent at 3-5 $mL \cdot min^{-1}$.

The remaining extract was loaded onto a calibrated gel permeation column (Biobeads SX-3), and eluted with a 1:1 dichloromethane:hexane solution. The fraction of

150-300 mL was collected, and concentrated prior to cleanup. The extract was applied to a calibrated Florisil® column, and eluted with hexane followed by 15:85 dichloromethane:hexane solution. The first and second fractions were collected together, and an aliquot of recovery standard solution, containing 13C-labelled PCB 153, was added prior to GC/MS analysis.

GC/MS analysis was carried out using a Finnigan INCOS 50 mass spectrometer equipped with a Varian 3400 GC, a CTC autosampler and a Prolab/Envirolink data system. Chromatographic separation of pesticides was achieved with a DB-5 chromatography column (60 m, 0.25 mm ID x 0.10 µm film). The MS was operated in the EI mode at unit mass resolution and in the MID (Multiple Ion Detection) mode acquiring two characteristic ions for each target analyte and internal standard. Concentrations of target analytes were calculated by comparing the area of the quantitation ion to that of the corresponding ^{13}C-labelled standard, and correcting for response factors. Concentrations of analytes were corrected based on the percent recovery of surrogate standards. Results were obtained for all six compounds of ΣDDT, and were expressed as nanograms of pesticide per gram wet weight of plant tissue ($ng \cdot g^{-1}$) (Appendix B). The analytical detection limit for this method was sample-specific and determined by surrogate recovery efficiency. Sample results at analytical detection limits were set to half of the sample-specific detection limit. The precision of analytical techniques was monitored with sample duplicates within an analysis batch.

4. Mammals

In total, 23 *S. parryi* livers were analysed by Axys Analytical Services Limited in Sidney, British Columbia. Analytical methods were identical to those described above

for plant tissue analysis, with a few exceptions. Following extraction, and prior to clean-up, the eluent was concentrated and sub-sampled for gravimetric lipid analysis. Results were obtained for all six compounds of ΣDDT, and were reported as nanograms of pesticide per gram wet weight of animal tissue ($ng \cdot g^{-1}$), and subsequently were lipid-normalized (Appendix B). The analytical detection limit for this method was sample-specific and determined by surrogate recovery efficiency. Sample results at analytical detection limits were set to half of the sample-specific detection limit. The precision of analytical techniques was monitored with sample duplicates within an analysis batch.

C. Data Analyses

The spatial distribution of ΣDDT compounds in surface soils was interpolated from 20 samples at the camp, and 135 samples at the station. Data obtained from discrete sample locations were used to create contour surfaces based on concentration. Spatial statistics were also used to assign appropriate soil ΣDDT concentrations and compositions to specific plants collected from the camp and the station. Data were analysed using Surfer® Contouring and 3D Surface Mapping (Version 7) software. From the variety of available gridding methods, Kriging was the approach selected for this study because for irregularly spaced data sets with less than 250 observations it provides better representation than other methods (Golden Software, Inc., 1999).

Organic carbon data and ΣDDT concentrations, data were log-transformed for linear regression analyses. Principal component analysis (PCA) using a covariance matrix was performed using the six compounds expressed as a percentage of the ΣDDT concentration. Data were transformed to a mean of zero and a standard deviation of one. PCA is a descriptive tool that is applied to reduce the dimensionality of a data set

consisting of a large number of interrelated variables, while retaining as much of the variability present in the data set as possible (Koprivnjak & Poissant, 1997). This reduction is achieved by transforming the data set into a new set of variables, the principal components (PCs), which are orthogonal (non-correlated), and arranged in decreasing order of importance.

An estimation of total daily intake (TDI) of ΣDDT by *S. parryi* was calculated using U.S. EPA and CCME exposure equations in Monte Carlo simulation software. In Monte Carlo simulation, a model is analysed in an iterative manner with varying input parameters, where uncertain variables are expressed as distributions rather than fixed values (CCME, 1997). Monte Carlo simulations were performed using @Risk (Version 4) software. The equation for calculation of TDI was as follows (CCME, 1997):

$$TDI = EDI_{soil} + EDI_{plant} \quad \text{(Equation 1)}$$

where,

$$EDI_{soil} = C_{soil} \times SIR \times F_{soil} \times BA \times AU / BW \quad \text{(Equation 2)}$$

and,

$$EDI_{plant} = C_{plant} \times PIR \times F_{plant} \times BA \times AU / BW \quad \text{(Equation 3).}$$

When calculating TDI, several conservative assumptions are made in modelling the soil-plant-herbivore pathway (CCME, 1997):

- 100% of the contaminant exposure of the herbivore originated from the ingestion of contaminated soil and food.
- The herbivore remained on the contaminated site 100% of the time.
- 100% of the food ingested by the herbivore was consumed from the contaminated site.

Accordingly, this study only considered exposure via the ingestion of contaminated soil or plants. TDI (Equation 1) was the sum of the estimated daily intake of contaminated soil (EDI_{soil}) (Equation 2) and the estimated daily intake of contaminated plants (EDI_{plant}) (Equation 3) (Table III-1). Exposure via contaminated drinking water, dermal contact with contaminated soil, and inhalation of contaminant vapour were not considered. The two latter assumptions were adjusted given published data on the life history of the study species. Given the variance in both the home range and dietary composition of *S. parryi*, exposure scenarios were calculated for two different diets from either the station or camp, as described below.

The EDI_{soil} was the product of the contaminant concentration in soil (C_{soil}), soil ingestion rate (SIR), fraction of soil in diet (F_{soil}), bioavailability of ingested contaminant (BA), usage of the contaminated area (AU), and divided by the body weight of the animal (BW). Analytical results for ΣDDT in soil were used as input variables for C_{soil}. The SIR was calculated according to the equation:

$$SIR = DMIR \times PSI \qquad \text{(Equation 4)}$$

SIR was the product of the dry matter intake rate (DMIR) and the soil ingestion proportion (PSI) (Equation 4). If a DMIR for an animal is unavailable, an allometric equation can be used to estimate the feeding rate of a mammal (F_M) using body weight (BW) according to the equation:

$$F_M = 0.0687 \times (BW)^{0.822} \qquad \text{(Equation 5)}$$

The next component of EDI_{soil}, F_{soil}, has a recognized default value of 0.07 (CCME, 1996). The BA of ingested contaminants was assumed to be 100%, or a value of one. AU is the ratio of the contaminated area to the home range of the animal in

Table III-1: Input parameters for calculation of TDI for *S. parryi*.

Variable	Description	Units	Source
TDI Equation			
TDI	Total daily intake of contaminant from all relevant pathways	$mg \cdot kg^{-1} \cdot bw \cdot day$	Calculated
EDI_{soil} Equation			
EDI_{soil}	Total daily intake of contaminant from soil pathway	$mg \cdot kg^{-1} \cdot bw \cdot day$	Calculated
C_{soil}	Contaminant concentration in soil in foraging area	$mg \cdot kg^{-1}$ dw	Field data
SIR	Soil ingestion rate on a dw basis	$mg \cdot day^{-1}$	Calculated
F_{soil}	Fraction of soil in diet	Unitless	Literature[1]
BA	Bioavailibility	Unitless	Estimated
AU	Area use as a ratio of the contaminated area to the home range	Unitless	Estimated
BW	Body weight	kg	Field Data
EDI_{plant} Equation			
EDI_{plant}	Total daily intake of contaminant from plant pathway	$mg \cdot kg^{-1} \cdot bw \cdot day$	Calculated
C_{plant}	Contaminant concentration in plants in foraging area	$mg \cdot kg^{-1}$ dw	Field data
PIR	Plant ingestion rate on a dw basis	Unitless	Calculated
F_{plant}	Fraction of plant material in diet	Unitless	Literature[2]
BA	Bioavailibility	Unitless	Estimated
AU	Area use as a ratio of the contaminated area to the home range	Unitless	Estimated
BW	Body weight	kg	Field data

1. (CCME, 1996)
2. (Batzli and Sabaski, 1980)

question. Since arctic ground squirrels can move up to 1 km within a day (Batzli & Sabaski, 1980), AU was given a value of one, or 100% usage of the contaminated area. Field data on actual BW were used for calculation.

The second component of TDI was EDI_{plant}, and was determined using a similar equation (Equation 3) to that for EDI_{soil} (Equation 4) the main difference being that C_{plant} was determined according to the equation:

$$C_{plant} = BAF \times C_{soil} \quad \text{(Equation 6)}$$

As a result, C_{plant} was based on site-specific BAFs and soil contaminant concentrations. Given the range of dietary preference for either monocotyledons, such as *Elymus* sp., and dicotyledons, *Salix* sp. (Batzli and Sabaski, 1980), exposures through two dietary scenarios were determined, where the diets contained either 100% *Salix* sp. or 100% *Elymus* sp. for F_{plant}. F_{plant} was based on a literature range of 64.5-93.3% of the total diet for *S. parryi* (Batzli & Sabaski, 1980).

Data distributions were determined with SYSTAT (Version 8) software and used as input for distribution specifications in @Risk. The TDI was the sum of the estimated daily exposure via soil ingestion (EDI_{soil}) and plant ingestion (EDI_{plant}).

BAFs for arctic ground squirrels were calculated using @Risk software. BAFs were determined through three pathways at either the station or camp: Salix sp.-animal, Elymus sp.-animal, and soil-animal. Values were expressed as: [kg dry soil or food]/[kg wet tissue (not lipid corrected)].

Following the calculation of TDI values from *Salix* sp. and *Elymus* sp. at the station and camp, dosages were used to estimate expected DDT liver burdens in arctic ground squirrels. Expected liver burdens were also calculated using the soil-leaf-mammal food chain BAF calculated by Jongbloed et al. (1996).

Toxic effects were investigated by comparing liver somatic indices (LSI) with ΣDDT concentrations in livers. Liver enlargement is one of the potential effects resulting from DDT exposure (Smith, 1991; ASTDR, 1994). LSI expresses liver size corrected for the body size for an organism. Liver somatic index (LSI) was calculated according to the equation:

$$LSI = \text{liver weight} / (\text{total weight} - \text{liver weight}) \quad \text{(Equation 7)}$$

IV. RESULTS

A. Soils and Sediments

Soils collected from both the station and the camp had similar physical characteristics. At both sites, surface soils collected near structures were typically sandy and contained less organic matter than soils collected further away from the building areas. At the camp, the organic carbon content of samples collected among the buildings ranged from 1.0-7.0%, whereas samples collected beyond the cabins, to the north-west, contained 13.6 and 25.4% organic carbon (n=5 and 2, respectively). Similarly, the organic carbon content in samples from within the station ranged from 1.5-9.7% (n=16). Samples with the highest organic carbon content, 13.3 and 18.5% (n=2), were located at the southeast edge of the built area.

Of the 135 soil samples collected from the station and 20 soil samples collected from the camp, detectable ΣDDT residues were found in 97 and 19 of the samples, respectively. The median concentration of ΣDDT in station soil samples was higher than in camp soils (Table IV-1). Concentrations of ΣDDT at the station ranged from 62-210,000 $ng \cdot g^{-1}$, and at the camp from 88-71,000 $ng \cdot g^{-1}$. Also, the contaminated area at the station was larger (approximately 4024 m^2) compared to the contaminated area at the camp (approximately 386 m^2). Contamination at the station was associated with site infrastructure, specifically former personnel accommodations (Figure IV-1). Similarly, contamination at the camp was localized to the area immediately surrounding the buildings (Figure IV-2).

Table IV-1: Concentrations of ΣDDT in surface soils collected from background locations, the station and the camp (n=2, 97 and 19, respectively). The applicable federal soil criterion is provided for comparison.

	Concentration ($ng \cdot g^{-1}$ dry weight)					
Location	***p,p'*-DDT**	***o,p'*-DDT**	***p,p'*-DDE**	***p,p'*-DDD**	***o,p'*-DDD**	**ΣDDT**
Background	ND	ND	ND	ND	ND	ND
Station						
Maximum	150000	45000	13000	8100	4000	210000
Median	700	120	75	100	42	1300
Minimum	ND	ND	ND	ND	ND	62
Camp						
Maximum	65000	6500	5000	2600	540	71000
Median	340	53	77	59	ND	540
Minimum	ND	ND	ND	ND	ND	88
Criterion						
CCME 1999[1]						700

Note: ND indicates a result below the analytical detection limit of 20 $ng \cdot g^{-1}$.

1. Canadian Council of Ministers of the Environment criterion for residential/agricultural land use.

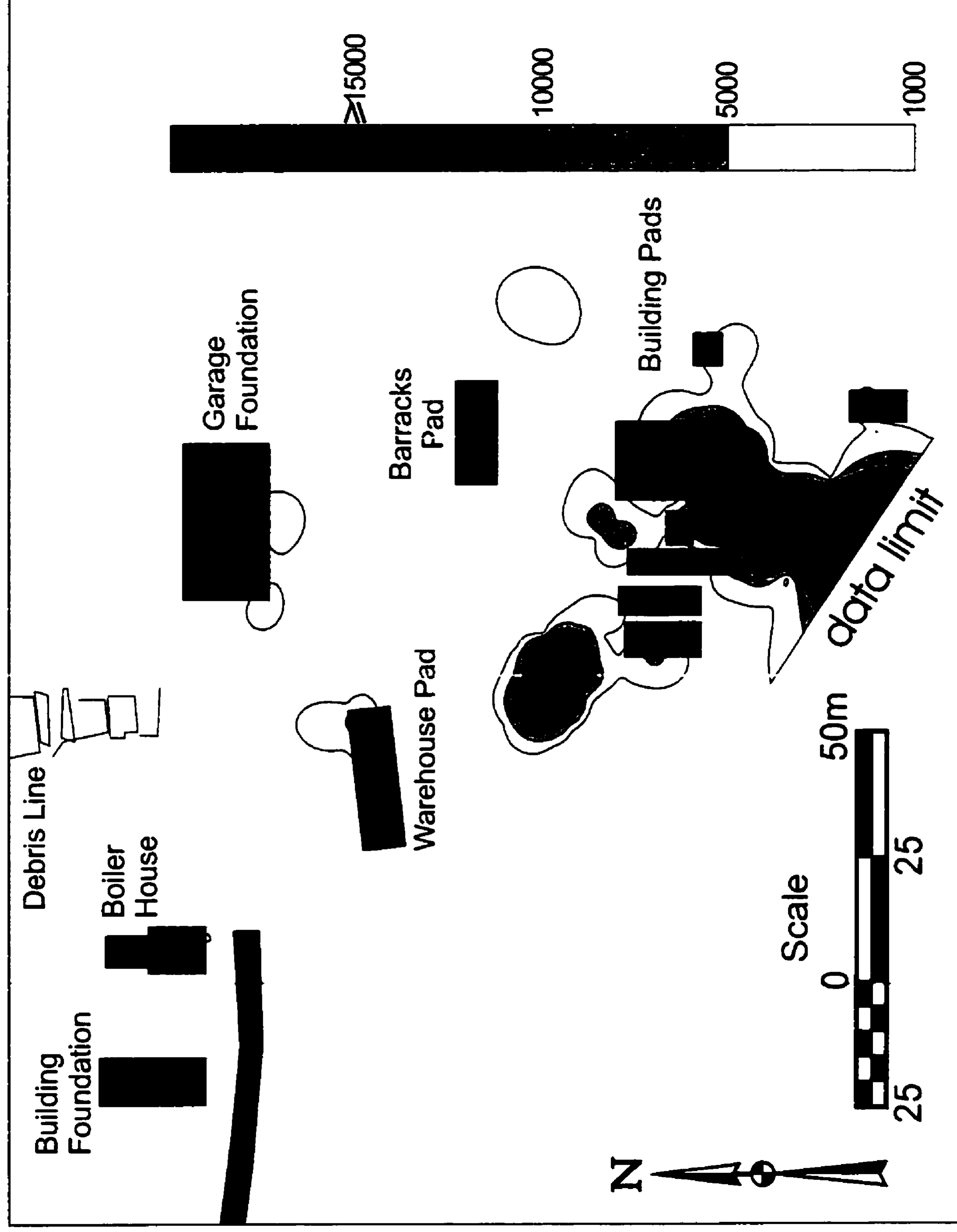

Figure IV-1: Spatial extent of ΣDDT contamination at the station. Distribution based on data for surface soils. Contaminant concentrations are in $ng \cdot g^{-1}$.

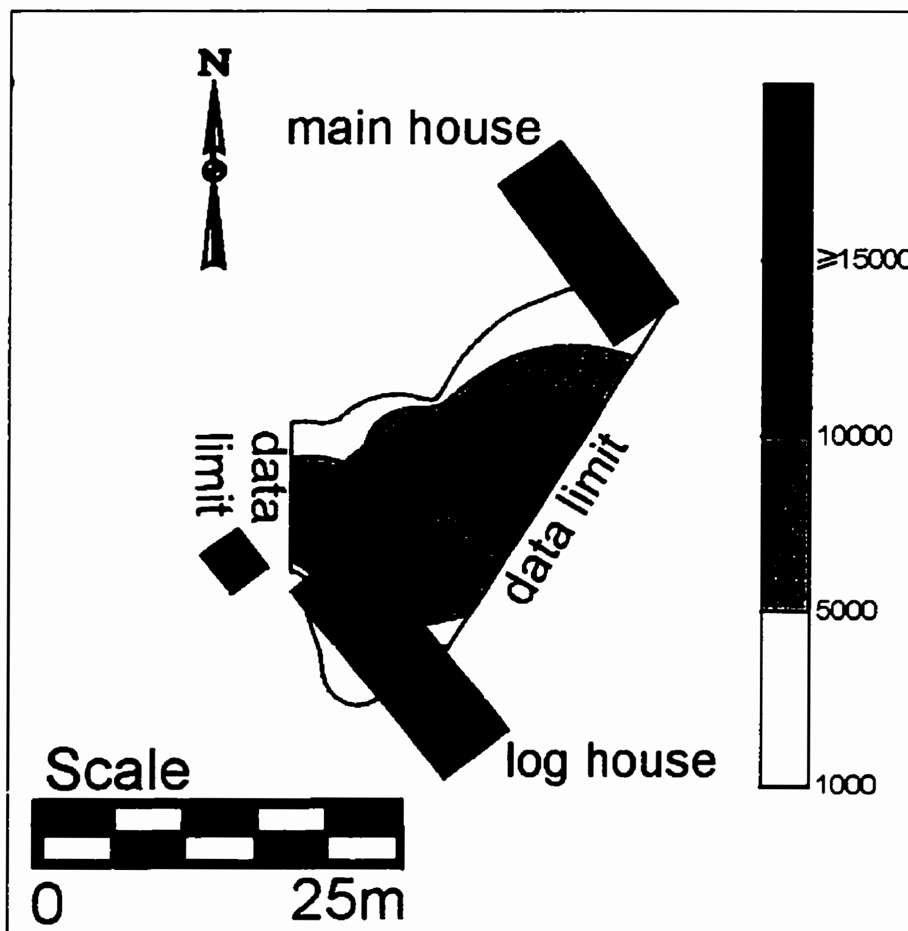

Figure IV-2: Spatial extent of ΣDDT contamination at the camp. Distribution based on data for surface soils. Contaminant concentrations are in $ng{\cdot}g^{-1}$.

Although the extent of contamination at the two sites differed, the relative composition of ΣDDT in samples was similar. In both station and camp soils, *p,p'*-DDT was the predominant compound and comprised 58.8 and 57.5% of detectable residues, respectively (Figure IV-3). At the station, the soil composition was 12.4% *o,p'*-DDT, 9.3% *p,p'*-DDE and 11.1% *p,p'*-DDD. Similarly, at the camp *o,p'*-DDT, *p,p'*-DDE and *p,p'*-DDD comprised 8.7, 16.3 and 10.5% of detectable residues, respectively. The

compound consistently found in the lowest percentage was *o,p'*-DDD at 3.9% at the station and 3.4% at the camp.

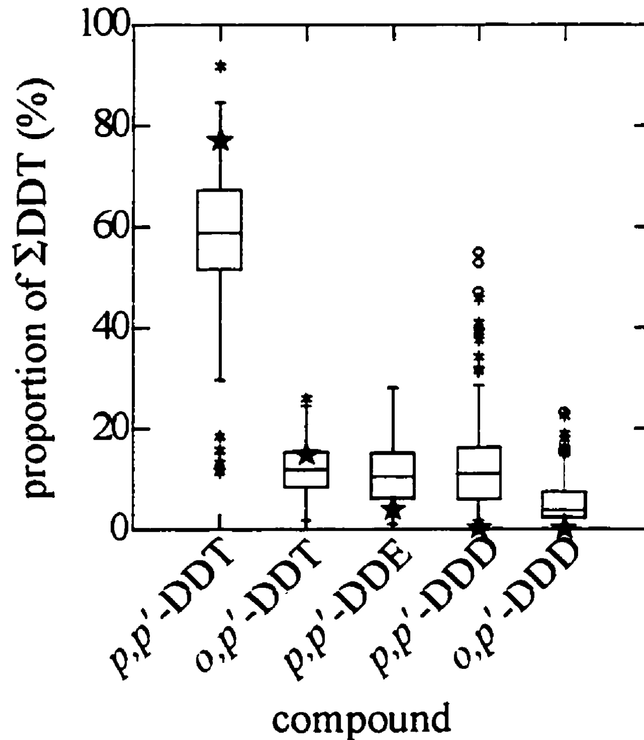

Figure IV-3: Relative ΣDDT composition of surface soil samples collected from both the camp and station (n=116). Compositions are expressed as the percent contribution of a specific compound to the total detectable concentration of ΣDDT. The composition of TG-DDT indicated by stars. The top and bottom of each box indicate the interquartile range and the horizontal line represents the median. Vertical lines indicate data within 1.5 interquartile ranges from either box edge. Asterisks represent data between 1.5-3 interquartile ranges and circles represent data beyond 3 interquartile ranges.

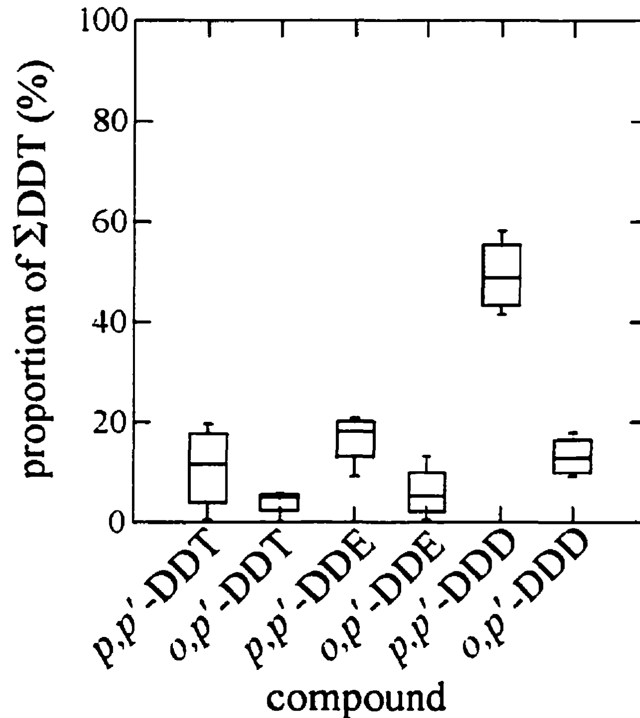

Figure IV-4: Relative ΣDDT composition of sediment samples collected from water bodies at both the camp and station (n=4). Compositions are expressed as the percent contribution of a specific compound to the total detectable concentration of ΣDDT.

The composition of ΣDDT in surface soils from the station and the camp was compared using PCA (Figure IV-5). Analysis of the two individual sites resulted in almost identical loading plots; no differences were observed between sites. Subsequent analysis of combined soil data, from both the sites, generated an almost identical plot, with no apparent grouping of samples on the basis of site (Figure IV-5). The first and second principal components explained 54% and 22% of the observed variance, respectively. The first component is representative of the ΣDDT concentration, whereas the second component is correlated to soil characteristics, such as percent organic carbon (Figure IV-5). One distinct and 2 less distinct groupings of samples were observed

(Figure IV-5). The large, distinct grouping at the left of the plot was influenced by the ΣDDT proportion comprised of *p,p'*-DDT and *o,p'*-DDT. Samples in this group were from highly contaminated areas located near either the buildings at the station, or the cabins at the camp. The grouping located at the upper-right was influenced by the proportion of *p,p'*-DDD and *o,p'*-DDD, whereas the grouping at the lower-right was influenced by the proportion of *p,p'*-DDE. Samples in these two groupings were generally from less contaminated areas located at the periphery of the two sites.

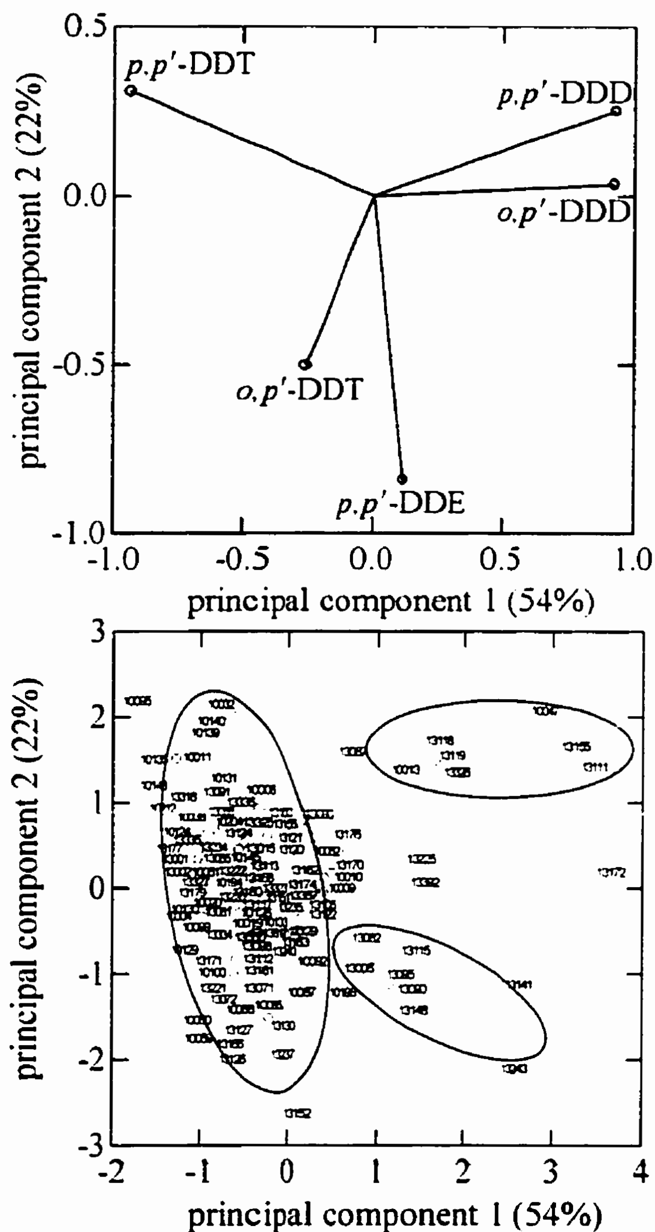

Figure IV-5: Principal components loading (top) and score (bottom) plots of the relative ΣDDT compositions of surface soil samples from both the camp and station areas (n=116). Values in parentheses indicate percent of total variance explained. For the first component, the regression relationship was described by: first principal component = 1.871 − 0.638($\log_{10}$ ΣDDT $ng \cdot g^{-1}$ soil dry weight) (linear regression, $r^2 = 0.332$, $p < 0.001$, $n = 114$). For the second component, the regression relationship was described by: second principal component = − 1.166 + 1.597($\log_{10}$ percent organic content) (linear regression, $r^2 = 0.348$, $p = 0.002$, $n = 24$).

B. Plants

Similar to surface soils, the ΣDDT concentration in plants differed between the station and the camp. Samples of *Elymus* sp. and *Salix* sp. collected from the station had a higher median ΣDDT concentration compared to samples collected from the camp (Table IV-2). The ΣDDT concentrations also differed between genera at both sites. At both sites, samples of *Salix* sp. had higher ΣDDT concentrations than *Elymus* sp. samples.

Table IV-2: Concentrations of ΣDDT in *Elymus* sp. and *Salix* sp. samples collected from background locations, the station and the camp (n=2, 13 and 3 for *Elymus* sp. and n=3, 16 and 3 for *Salix* sp., respectively).

	Concentration (ng·g^{-1} dry weight)						
	p,p'-DDT	*o,p'*-DDT	*p,p'*-DDE	*o,p'*-DDE	*p,p'*-DDD	*o,p'*-DDD	ΣDDT
			***Elymus* sp.**				
Background	ND	ND	ND	ND	ND	ND	ND
Station							
Maximum	920	200	260	4.0	108	52	1500
Median	200	58	74	0.74	5.2	3.5	350
Minimum	1.0	0.44	1.0	0.020	0.17	0.080	3.2
Camp							
Maximum	72	20	34	1.2	4.6	0.86	130
Median	65	6.2	22	1.0	2.1	0.77	99
Minimum	30	4.3	8.6	0.14	1.4	0.48	46
			***Salix* sp.**				
Background	0.14	0.03	0.06	ND	ND	ND	0.24
Station							
Maximum	5800	1900	1800	34	340	140	10000
Median	310	56	75	1.6	26	11	470
Minimum	7.0	0.86	3.3	0.24	0.30	0.27	12
Camp							
Maximum	130	16	81	0.83	9.2	2.5	230
Median	65	9.4	36	0.63	7.8	1.5	120
Minimum	64	7.1	20	0.19	2.0	0.50	100

Note: ND indicates a result below the sample specific detection limit for specified compound.

Although the ΣDDT concentrations differed between genera, the relative compositions of ΣDDT in plant samples were similar between the station and the camp, as well as genera (Figure IV-6). The composition of ΣDDT in plants was similar to that in soils (Figure IV-6 and IV-3, respectively). The relative contribution of each compound was identical between genera and was predominantly *p,p'*-DDT, *p,p'*-DDE, and *o,p'*-DDT. For *Salix* sp. samples, the composition of ΣDDT was 58.1% *p,p'*-DDT, 21.4% *p,p'*-DDE, 11.2% *o,p'*-DDT, 4.1% *p,p'*-DDD, 1.4% *o,p'*-DDD and 0.3% *o,p'*-DDE. For *Elymus* sp. samples, the composition of ΣDDT was 64.3% *p,p'*-DDT, 18.1% *p,p'*-DDE, 12.0% *o,p'*-DDT, 3.2% *p,p'*-DDD, 1.1% *o,p'*-DDD and 0.3% *o,p'*-DDE.

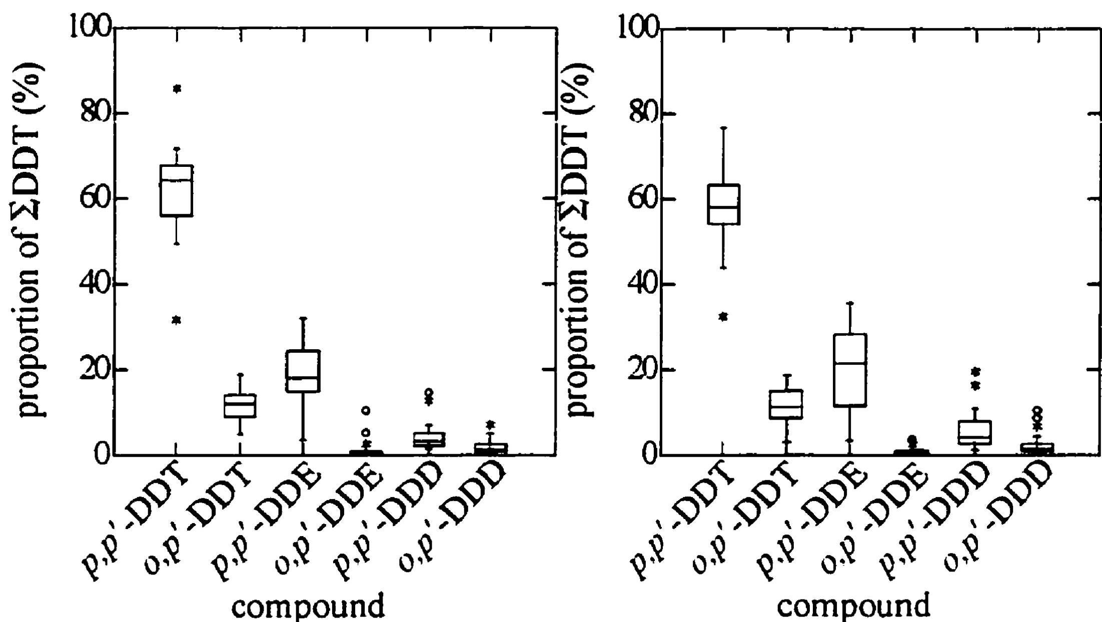

Figure IV-6: Relative ΣDDT composition of *Elymus* sp. (left) and *Salix* sp. (right) samples collected from both the camp and station (n=16 and 19, respectively). Compositions are expressed as the percent contribution of the specific compound to the total detectable concentration of ΣDDT.

The composition of ΣDDT in plants from the station and the camp was also compared using PCA. Analysis of pooled plant data from both sites (Figure IV-7) generated an almost identical result to the individual sites. In general, results for plants were similar to that of soils. The first and second principal components explained 40% and 32% of the observed variance, respectively. As with the soil samples, the first component correlated to a ΣDDT concentration gradient (Figure IV-7). Given the location of samples along the second component, this component appears to reflect soil characteristics that change with distance from the site. Similar to the PCA for soil samples, one distinct and two less distinct groupings of plant samples were observed (Figure IV-7). The distinct grouping at the left of the plot was influenced by the ΣDDT proportion of *p,p'*-DDT. Samples in this group were collected from highly contaminated areas and were located near either the buildings at the station, or the cabins at the camp. The grouping located at the upper-right was influenced by the proportion of *p,p'*-DDD and *o,p'*-DDD, whereas the grouping at the lower-right was influenced by the proportion of *p,p'*-DDE. Both latter groups contained samples collected from less contaminated areas located at the periphery of the two sites.

The similarity between soil and plant ΣDDT composition was examined by calculating the compound-specific bioaccumulation factors (BAFs). Calculated BAFs for plants using interpolated soil ΣDDT concentrations, were similar within genera (Table IV-3). No distinct preference for the uptake of a particular compound was observed. For example, BAFs for *Elymus* sp. ranged from 0.010 for *p,p'*-DDD to 0.051 for *p,p'*-DDE and BAFs for *Salix* sp. ranged from 0.027 for *o,p'*-DDD to 0.177 for *p,p'*-DDE. In general, the ΣDDT concentration and composition for both genera reflected the

concentration and composition of contaminants in the soil from which they were collected.

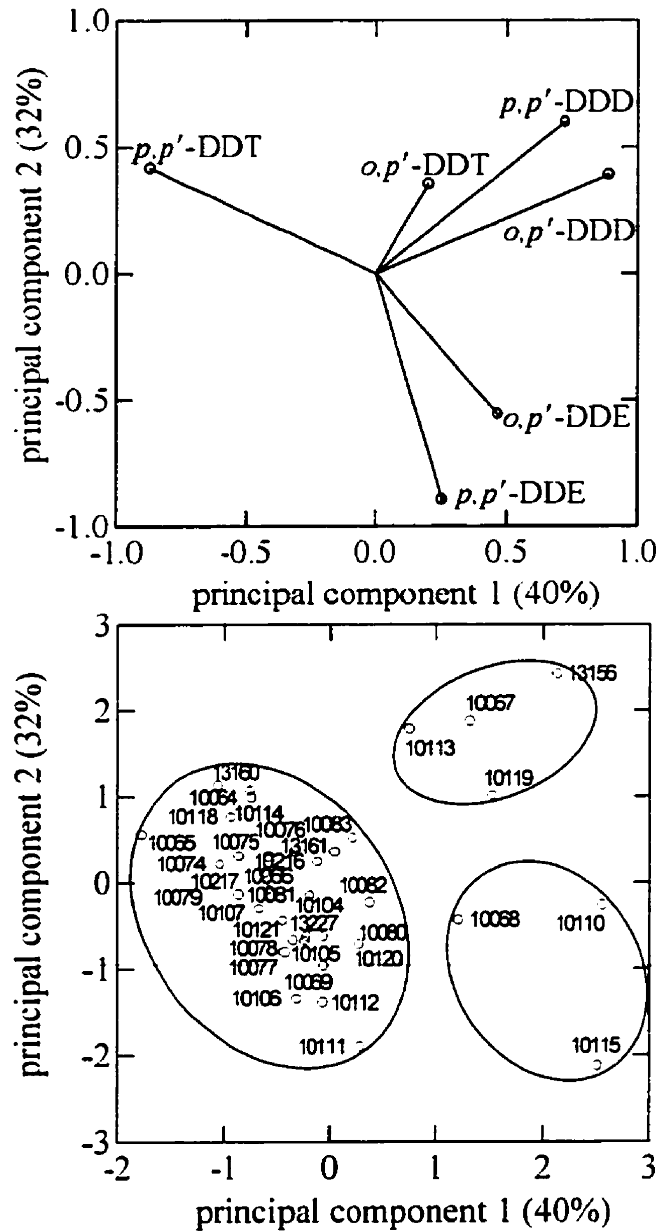

Figure IV-7: Principal components loading (top) and score (bottom) plots of the relative ΣDDT compositions of *Elymus* sp. and *Salix* sp. samples from the station and camp areas (n 35). Values in parentheses indicate percent of total variance explained.

Table IV-3: Compound-specific bioaccumulation factors for *Elymus* sp. and *Salix* sp. (n=16 and 19, respectively). Factors are expressed on both a plant dry weight to soil dry weight and a plant wet weight to soil dry weight basis

	Bioaccumulation Factor					
	***p,p'*-DDT**	***o,p'*-DDT**	***p,p'*-DDE**	***p,p'*-DDD**	***o,p'*-DDD**	**ΣDDT**
***Elymus* sp.**						
Dry: Dry	0.025	0.013	0.051	0.010	0.014	0.021
Wet: Dry	0.007	0.004	0.015	0.002	0.003	0.006
***Salix* sp.**						
Dry: Dry	0.066	0.065	0.177	0.031	0.027	0.076
Wet: Dry	0.027	0.026	0.079	0.025	0.012	0.031

For all compounds, *Salix* sp. samples had consistently greater BAFs than *Elymus* sp. samples. The differences in BAFs between genera appear significant, but they are likely an artefact of differential lipid content between genera. Although lipid data were unavailable for plant samples in this study, percent lipid content for plants of comparable taxonomy (*Salix* sp. and various grasses) were available from a study site at Saglek, Labrador. Samples of *Salix arctica* and *Salix herbacea* contained a mean of 1.7% lipid, and samples of various grasses contained a mean of 0.37% lipid (n=8 and 3, respectively) (Z.A Kuzyk, personal communication). Using lipid normalised plant concentrations, *p,p'*-DDT BAFs of 1.6 for *Salix* sp. and 1.9 for *Elymus* sp. were calculated. Hence, correction of BAFs on the basis of lipid content produces similar BAFs between genera.

C. Mammals

Physical characteristics of *S. parryi* collected from the station, camp, access road and background sites were similar. Slight differences in physical characteristics between the sexes were observed (Table IV-4).

Table IV-4: Comparison of physical characteristics between sexes of *S. parryi* collected from all areas. Values are expressed as means (± standard deviation).

Sex	Number[1]	Length (cm)	Mass (kg)	Liver Mass (g wet weight)	Liver Lipid (%)
M	11	26.5 (3.5)	0.61 (0.22)	32.8 (15.6)	4.6 (1.1)
F	11	24.9 (3.4)	0.48 (0.24)	26.2 (12.9)	4.0 (0.67)

1. Sex, length, and mass data unavailable for one sample.

Of the 23 *S. parryi* samples collected, only two failed to contain detectable ΣDDT concentrations in liver tissue. One came from a background location, and the other was a sample collected along the access road between the station and the camp. Liver concentrations of ΣDDT were associated with proximity of the squirrel to contaminated areas (Table IV-5). The highest ΣDDT concentration of 4300 $ng \cdot g^{-1}$ was from a station specimen. In comparison, the highest concentration at the camp was 1400 $ng \cdot g^{-1}$, and the highest concentration along the road was 33 $ng \cdot g^{-1}$.

Table IV-5: Concentrations of ΣDDT in *S. parryi* collected from a background location, the station, the camp and the access road (n=2, 11, 3, and 7, respectively).

	Concentration ($ng \cdot g^{-1}$)						
	***p,p'*-DDT**	***o,p'*-DDT**	***p,p'*-DDE**	***o,p'*-DDE**	***p,p'*-DDD**	***o,p'*-DDD**	**ΣDDT**
Background	ND	ND	4.5	ND	ND	ND	4.5
Station							
Maximum	10	17	1500	1.7	2800	29	4300
Median	0.74	1.0	78	0.075	62	0.32	140
Minimum	0.08	0.12	0.69	ND	1.1	ND	2.3
Camp							
Maximum	4.5	3.3	290	3.9	1100	1.7	1400
Median	1.4	2.8	140	0.90	280	1.0	430
Minimum	1.2	0.90	1.2	0.081	2.6	0.50	7.3
Road							
Maximum	1.1	1.1	8.4	0.89	21	0.44	33
Median	0.12	0.19	3.3	0.07	5.5	0.054	11
Minimum	0.06	0.076	0.95	ND	1.9	ND	3.1

Note: ND indicates a result below the sample specific detection limit for specified compound.

The relative ΣDDT composition in *S. parryi* samples was notably different from those in soils and plants. The compounds *p,p'*-DDD and *p,p'*-DDE were the most predominant and comprised 61.8 and 32.5% of the ΣDDT composition of samples, respectively (Figure IV-8). The contribution of remaining compounds was 1.2% *o,p'*-DDT, 0.9% *p,p'*-DDT, 0.6% *o,p'*-DDE, and 0.4% *o,p'*-DDD.

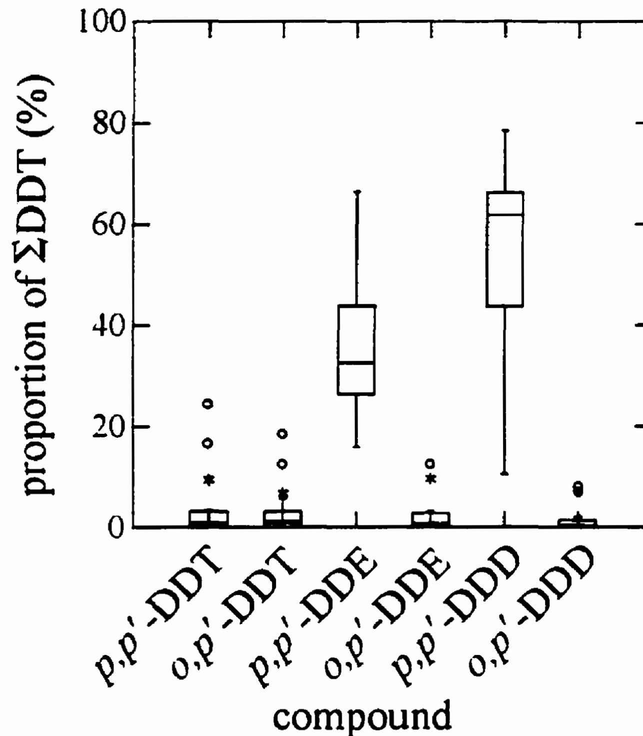

Figure IV-8: Relative ΣDDT composition of *S. parryi* collected from all areas (n=21). Compositions are expressed as the percent contribution of a specific compound to the total detectable concentration of ΣDDT.

A PCA of the ΣDDT composition in liver tissues was initially conducted using all samples with detectable residues. Using this method, principal component 1 explained 72% of the observed variance. Examination of the principal component scores indicated that two samples were largely responsible for the result. As the two samples contained the lowest ΣDDT concentrations of all samples, they were judged to have an undue influence and were removed from subsequent analysis. Following the removal of these two samples with ΣDDT concentrations below analytical detection limits, the variance explained by principal component 1 was 58% and by principal component 2 was 32% (Figure IV-9). Similar to the results obtained for surface soils and plants, principal component 1 was correlated to ΣDDT concentration. In general, samples with high

ΣDDT concentrations were dominated by large proportions of *p,p'*-DDD and *p,p'*-DDE. Groupings of samples along principal component 1 appear to be associated with proximity to contaminated areas. Samples at the right are somewhat removed from the immediate vicinity of the site, whereas the grouping at the left contains almost all of the samples collected closest to either the station or camp area. Principal component 2 correlated to physical characteristics of the animal (Figure IV-9). Length, body mass and liver mass were all positively correlated with the proportion of *p,p'*-DDE in the liver. No relationships or grouping on the basis of sex were observed.

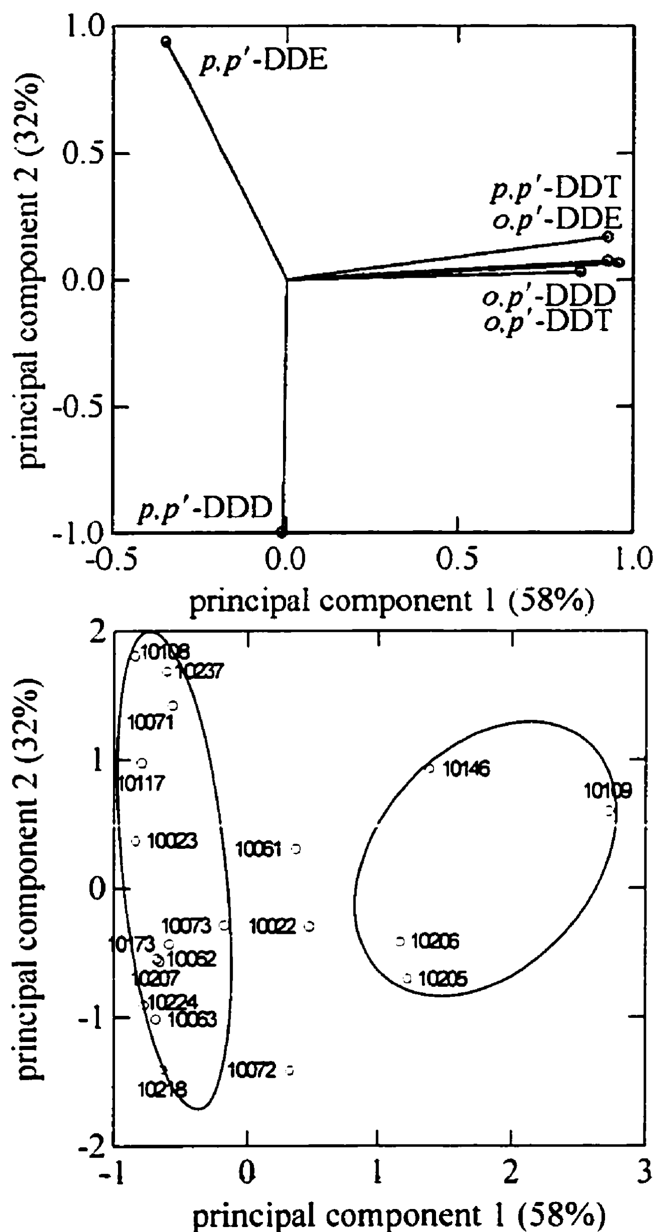

Figure IV-9: Principal components loading (top) and score (bottom) plots of the relative ΣDDT compositions of *S. parryi* samples from all areas (n=21). Values in parentheses indicate percent of total variance explained. For the first principal component: principal component 1 = -0.079 – 0.617($\log_{10}$ ΣDDT $ng \cdot g^{-1}$ lipid) (linear regression, r^2=0.481, p<0.001, n=19). For principal component 2: principal component 2 = -1.374 + 0.044(liver mass g wet weight) (linear regression, r^2=0.395, p<0.005, n=19).

BAFs were calculated for arctic ground squirrels collected at the station and camp (Table IV-6). BAFs for plant tissue-animal tissue pathways were of similar magnitude, and resulted in a mean of 0.0081 between the station and camp. In comparison, BAFs for soil-animal tissue pathway were much lower, and had a mean of 0.00023 between the station and camp.

Table IV-6: *p,p'*-DDT BAFs for arctic ground squirrels. BAFs were calculated using wet weight tissue concentrations (not lipid corrected) and dry weight plant and soil concentrations. Liver concentrations of animals at the station and camp (n=7 and 3, respectively) were divided by the range of plant or soil concentrations at either the station or camp.

Pathway	BAF Mean (SD)	BAF Range
Station		
Salix sp.-animal	0.0086(0.028)	3.6×10^{-6}-0.40
Elymus sp.-animal	0.016(0.057)	4.5×10^{-6}-0.98
soil-animal	0.00036(00013)	4.7×10^{-7}-0.026
Camp		
Salix sp.-animal	0.0027(0.0083)	4.0×10^{-6}-0.10
Elymus sp.-animal	0.0049(0.016)	4.8×10^{-6}-0.25
soil-animal	0.00010(0.00021)	9.3×10^{-7}-0.0032

Two exposure scenarios were used to derive the most probable total daily intake (TDI) for ground squirrels exposed to contamination at the station. Calculation of the TDI via two exposure pathways was as follows:

$$TDI = EDI_{soil} + EDI_{plant} \quad \text{(Equation 1)}$$

where,

$$EDI_{soil} = C_{soil} \times SIR \times F_{soil} \times BA \times AU / BW \quad \text{(Equation 2)}$$

and,

$$EDI_{plant} = C_{plant} \times PIR \times F_{plant} \times BA \times AU / BW \quad \text{(Equation 3)}$$

Field measured variables and corresponding distributions were available for C_{soil}, BW, $BAF_{Salix\ sp.}$, $BAF_{Elymus\ sp.}$, and were used to calculate values for $C_{Salix\ sp.}$, $C_{Elymus\ sp.}$, DMIR, SIR, PIR and FIR. Under no scenario did the most probable TDI exceed the NOEL (Table IV-7). Despite differences in BAFs between genera, the most probable TDIs were similar between the two dietary compositions. Sensitivity analysis identified that C_{soil} was the most influential variable in determining TDI.

Table IV-7: Monte Carlo simulation results for calculation of TDI of DDT through the ingestion of either *Salix* sp. or *Elymus* sp. at the station or camp. Probability of exceeding NOEL for each scenario is also provided for each scenario.

Diet	Mean TDI (mg·kg bw·day^{-1})	TDI Range (mg·kg bw·day^{-1})	Probability of TDI ≤ NOEL[1]
Station			
100% *Salix* sp.	0.055(0.27)	6.3×10^{-5}-7.2	0.98
100% *Elymus* sp.	0.024(0.085)	7.8×10^{-5}-2.2	1.00
Camp			
100% *Salix* sp.	0.053(0.13)	1.2×10^{-4}-2.3	0.97
100% *Elymus* sp.	0.022(0.048)	1.2×10^{-4}-0.79	1.00

1. NOEL for DDT of 0.37 mg·kg^{-1} bw·day^{-1}.

Calculated TDI values were used to generate expected liver burden concentrations given certain assumptions. Since arctic ground squirrels spend 8-10 months in hibernation (Yukon Department of Renewable Resources, 2000), the ingested dose was based on 90 days of activity during which exposure could occur. Also, the depuration of DDT from animals was conservatively assumed to be zero. Expected liver burdens were much higher than observed tissue concentrations (Table IV-8). Since ingested *p,p'*-DDT could be converted to either DDE or DDD, observed ΣDDT concentrations were also

compared. Even when compared on the basis of ΣDDT concentration, expected tissue concentrations were 100-200 fold greater than observed.

Table IV-8: Expected *p,p'*-DDT and ΣDDT liver burdens based on calculated TDI values for different diets at the station and camp.

Diet	*p,p'*-DDT ($ng \cdot g^{-1}$)[1]		*p,p'*-DDT ($ng \cdot g^{-1}$)[1]		ΣDDT ($ng \cdot g^{-1}$)[1]	
	Expected Mean (SD)	**Expected Range**	**Observed Mean (SD)**	**Observed Range**	**Observed Mean (SD)**	**Observed Range**
Station			0.18(200)	0.034-0.470	74(54)	10-150
Salix sp.	4100(12000)	4.4-140000				
Elymus sp.	2200(6300)	6.3-87000				
Camp			0.095(0.065)	0.055-0.170	23(27)	0.37-54
Salix sp.	4900(17000)	8.0-260000				
Elymus sp.	2100(3700)	9.1-47000				

1. Concentrations expressed on a wet weight basis without lipid correction.

Expected liver burdens were also calculated using another method. A soil-leaf-mammal BAF has been determined by Jongbloed et al. (1996) as 0.067 following a review of available data on various plants, and rodents. The final value of 0.067 was the median of reviewed data. Using this method, expected liver burdens were closer to observed concentrations, but again expected values were approximately 100 fold greater than observed values. Given the potential for DDT to become DDE or DDD once ingested, expected concentrations are best compared with observed ΣDDT concentrations.

Table IV-9: Expected *p,p'*-DDT and ΣDDT liver burdens based on soil-leaf-mammal pathway BAF of 0.067[1].

Location	*p,p'*-DDT ($ng \cdot g^{-1}$)[2]		*p,p'*-DDT ($ng \cdot g^{-1}$)[2]		ΣDDT ($ng \cdot g^{-1}$)[2]	
	Expected Mean (SD)	Expected Range	Observed Mean (SD)	Observed Range	Observed Mean (SD)	Observed Range
Station	330(780)	1.1-9200	0.18(200)	0.034-0.470	74(54)	10-150
Camp	400(610)	1.9-4100	0.095(0.065)	0.055-0.170	23(27)	0.37-54

1. Soil-leaf-mammal pathway BAF determined using geometric means of reviewed literature.
2. Concentrations expressed on a wet weight basis without lipid correction.

Potential physiological effects were investigated using calculated liver somatic indices (LSI) and lipid normalized contaminant concentration. LSI was found to increase with increasing concentrations of the ΣDDT in the liver (Figure IV-11). The relationships between LSI and contaminant concentration were similar between the sexes.

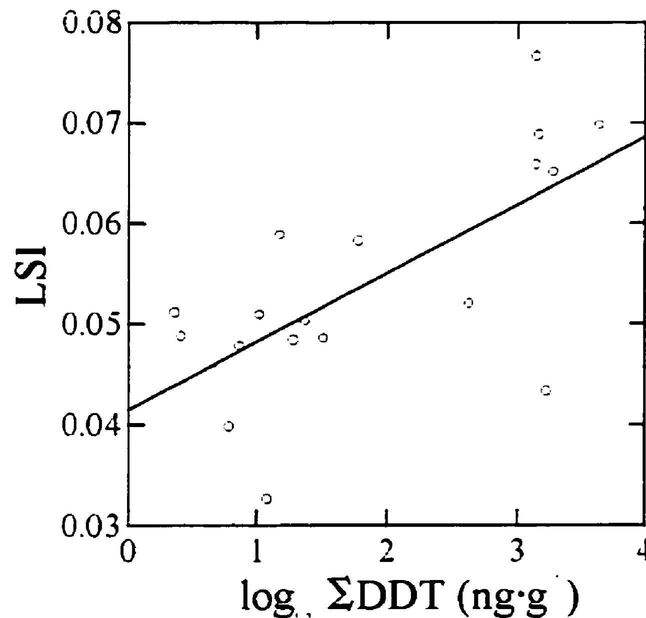

Figure IV-10: Relationship between LSI, and ΣDDT for both sexes of *S. parryi* from all locations. LSI was found to increase with increasing concentrations of ΣDDT. For ΣDDT: LSI = 0.041 + 0.007 (log_{10} ΣDDT $ng \cdot g^{-1}$) (linear regression, $r^2 = 0.450$, $p = 0.002$, $n = 18$).

V. DISCUSSION

A. Soils and Sediments

Soils contaminated with ΣDDT at the LORAN station and the reindeer herding camp differed both in spatial extent and concentration. At the station, ΣDDT was at higher concentrations and distributed over a larger area in comparison to the camp. According to applicable federal guidelines, the criterion for ΣDDT in soil is 700 $ng \cdot g^{-1}$ (CCME, 1999). Using this definition, a contaminated area of approximately 4024 m^2 was identified at the station, and approximately 386 m^2 at the camp. The highest contamination at the camp was located central to the remaining building and building foundations, and at the station the highest contamination was found amongst locations of former personnel and operational buildings.

The global distillation hypothesis explains the ubiquitous, regional presence of pollutants in the Arctic with little to no historical local use. Although ΣDDT compounds are candidates for long-range transport (Wania & Mackay, 1993; INAC, 1997), the contribution of this transport mechanism to the situation observed at Kittigazuit is negligible because an abrupt transition exists between soil contaminant levels at the sites, and samples collected immediately off-site. For example, the median ΣDDT concentration was 1300 $ng \cdot g^{-1}$ at the station, and 540 $ng \cdot g^{-1}$ at the camp. In comparison, results for soil samples considered representative of background conditions were below the analytical detection limit of 20 $ng \cdot g^{-1}$. Also, the ΣDDT composition at the sites was dominated by *p,p'*-DDT. This pattern is inconsistent with a long-range transport scenario in which *p,p'*-DDE would be the dominant compound due to its higher volatility. The

spatial distribution of ΣDDT contamination at the station and camp clearly indicates a legacy of historical use.

Using climate data from Tuktoyaktuk, the role of the pesticide use for mosquito control, and the record of daily spraying, the duration of DDT use at the station can be estimated. The station was operational from June 1948 to March 1950 (Hart & Cockney, 1999), and pesticide application presumably occurred only during July and August when daily mean temperatures are 11.0°C and 9.1°C, respectively (EC, 1998b). Hence, DDT contamination at the station was the result of only 6 months of use approximately 50 years ago.

No historical records documenting the use of DDT pesticides at the reindeer herding camp were found; it can, however, be assumed that DDT use at the camp mimicked that at the station to some degree. Lower ΣDDT concentrations, and the smaller area of contamination at the camp, probably indicates less frequent use, and/or a different mechanism of application compared to the station.

It should be noted that this study describes the current concentration and composition of ΣDDT in soils, and not the conditions immediately following closure of the site. During the intervening 50-year period, a variety of environmental processes have resulted in the current ΣDDT distribution. To understand the current status, the potential contribution of each process must be addressed. As noted in Chapter II, potential environmental fates, following the release of a pesticide, include wind drift, runoff, leaching, root uptake, sorption, volatilisation, chemical degradation, photodecomposition, and microbial degradation (Aislabie & Lloyd, 1995). Over time, these processes can result in decreased pesticide concentrations and/or altered pesticide composition. The

relative importance of each process depends on the characteristics of the pesticide, as well as of the environment itself.

Environmental fates of little relevance to this study are wind drift, runoff, leaching, root uptake, sorption, and volatilisation. Loss via wind drift refers to the loss of pesticide immediately following release such that the pesticide fails to reach its target area. Pesticide lost in this manner during application is irrelevant given the focus of this study on long-term rather than immediate, residues in soil.

Runoff can account for the loss of ΣDDT from a treated area via transport in solution or adhered to particulates in water runoff. At Kittigazuit, loss of ΣDDT in runoff is unlikely given the moderate topography, vegetated soils, low precipitation, and the relative insolubility of ΣDDT compounds in water. Also, since the physical extents of the contaminated areas were identified, and were coincident with areas of use, it does not appear as though the residues have been transported from either the station or the camp to a significant degree.

Leaching of ΣDDT compounds to lower soil horizons is generally considered insignificant given their low water solubility. For example, Boul et al. (1994), rejected this process as an explanation for the reduction in residue concentrations with time during a 40 year study of agricultural plots in New Zealand. Others (Morrison & Newell, 1999) have confirmed that DDT is generally not leached through soil profiles. Situations where residues have been located at depth have been explained by site-specific mechanisms. At a pesticide mixing facility in Fresno, California, DDT was found below surface soils due to dissolution into a solvent prior to its release (Morrison & Newell, 1999). Locations in the station at Kittigazuit where ΣDDT was found at depths of 60 cm were likely

transported in a similar manner. As noted in the station diary, reference is made to a pesticide solution into which cloth was soaked and used on windowsills (Hart & Cockney, 1999). Locations where this solution was either stored or dumped probably transported ΣDDT to greater depths.

Residue uptake and incorporation into plant tissue can account for pesticide loss from soil. The retention of ΣDDT residues in the uppermost soil horizons places them in the rhizosphere of most plants, thereby subjecting them to uptake. This fate is typical of agricultural situations where compounds, which have been incorporated into plants, can potentially leave the site during harvest, or through ingestion by animals. Although this study indicates ΣDDT uptake by both *Elymus* sp. and *Salix* sp., soil residue loss via root uptake is insignificant. This mechanism of loss has been dismissed in other long-term studies (Boul et al., 1994).

Concentrations of pesticides in soil can appear to decrease with time as compounds become bound to soil particles. Bound residues are essentially unavailable to biological systems, and are effectively lost from the environmental system. Since this study identified residues in plants and animals, ΣDDT compounds in soil are not completely bound and are at least partially bioavailable.

Under certain conditions, volatile loss can be the predominant fate of DDT pesticides, and in extreme circumstances, can alone account for the loss of DDT from soil. A field test of the rate of disappearance of DDT from soil near Lake Nakuru, Kenya, found that DDT sublimed directly without prior degradation to DDE (Sleicher & Hopcraft, 1984). In India, high soil temperature, intense sunlight and humidity were identified as the major factors responsible for dissipation by volatilisation (Samuel & Pillai, 1989). Temperature

has been shown to have a large influence on volatile loss. Using [^{14}C]- *p,p'*-DDT in sandy loam soil, loss through volatilisation increased five-fold when the temperature changed from 15 to 45°C (Samuel & Pillai, 1989). The colder temperatures at Kittigazuit would have the opposite effect, and result in decreased DDT loss via volatilisation.

Given the limited importance of wind drift, runoff, leaching, root uptake, sorption, and volatilisation in ΣDDT loss at Kittigazuit, the soil concentrations found in this study were likely similar to those immediately following closure of the site. As the initial ΣDDT concentration in soil was unavailable, and given the wide range of published half-lives for DDT, quantifying the actual extent of degradation on the basis of current concentrations is not possible. Insight regarding the qualitative extent of degradation is, however, provided by the relative composition of ΣDDT compounds in soil.

The composition of ΣDDT residues in soils from this study was inconsistent with patterns observed in temperate and tropical studies (Table V-1). In temperate and tropical environments, the chemical compositions are altered relatively rapidly, leaving little of the initial DDT pesticide.

At Fort Churchill, in a similar environment to that of Kittigazuit, almost equal proportions of the degradation products, DDE and DDD accompanied a predominance of the primary active ingredient, DDT. At Kittigazuit, the two possible degradation products of *p,p'*-DDT were found in equal amounts. Under reducing conditions, DDD is reported to be the predominant biotransformation product, whereas under oxidising conditions, DDE is the predominant biotransformation product (Gambrell et al., 1989). Although the conditions favouring the production of either DDE or DDD differ, finding equal proportions of DDE or DDD is not uncommon, particularly in temperate climates.

Soils in forests sprayed aerially with DDT in 1958-1967 were sampled for persistence of residues at subsequent intervals and most recently in 1993 (Dimond & Owen, 1996) at which time the biotransformation compounds DDE and DDD each comprised approximately a third of the total residues.

Table V-1: Initial and final ΣDDT composition in various soil studies. Studies are ranked according to increasing latitude.

Location	Time (years)	Initial Composition	Final Composition	Reference
Kenya	3	~80% *p,p'*-DDT	Direct sublimation of DDT without prior degradation to DDE, or DDD	Sleicher & Hopcraft, 1984
India	1	*p,p'*-DDT	29% *p,p'*-DDT	Singh & Agarwal, 1995
India	½	*p,p'*-DDT	61.3% *p,p'*-DDT	Samuel & Pillai, 1991
California, USA	23	TG-DDT	72-81% *p,p'*-DDE	Spencer et al., 1996
New Zealand	45	TG-DDT	54-71% *p,p'*-DDE, *p,p'*-DDT remainder with trace *p,p'*-DDD and *o,p'*-DDT depending on treatment	Boul et al., 1994
Maine, USA	26	TG-DDT	39% DDT, 31% DDE and 30% DDD	Dimond & Owen, 1996
Fort Churchill, Manitoba	3	TG-DDT	67.8% DDT, 19.6% DDE and 12.6% DDD	Brown & Brown, 1970

When chemical degradation, photodecomposition, or microbial degradation determines the fate of a pesticide, the result is usually an altered chemical composition of the pesticide. Compared with the composition of TG-DDT, the current composition of ΣDDT in surface soils at Kittigazuit indicates only a slight alteration since closure of the site.

The degradation pathways from DDT to either DDE or DDD are not mutually exclusive, and both products can be produced simultaneously. Though no obvious preference for either product was observed, principal components analysis (PCA)

revealed that samples obtained some distance from formerly, high traffic areas at the camp and station largely consisted of the degradation products of *p,p'*-DDD and *o,p'*-DDD. Samples with predominantly *p,p'*-DDE were from disturbed areas. Differences in soil conditions provide a likely explanation for the observed spatial separation of areas with DDD and areas with DDE.

Historical evidence from three sources indicates that the land immediately occupied by the building areas was disturbed by operations at the station. The area occupied by the station building was cleared of vegetation, and mechanically disrupted by machinery, as noted in archival photographs of the site during operation (Hart & Cockney, 1999). Samples containing predominantly DDE were located near such disturbed areas, where conditions were sandy, and lower in organic soil content. Samples containing predominantly DDD were located further away from disturbed areas, where organic soil contents were higher, and were likely representative of original soil conditions.

The different conditions appeared to favour different degradation products. Soil conditions can greatly influence degradation, and the observed pattern is consistent with other research. The sandy, and as a result, well-aerated conditions from disturbed areas favour the degradation of DDT to DDE. Under dry conditions, DDE tends to be the predominant residue (Childs & Boul, 1995 in Aislabie et al., 1997). In comparison, the more organic conditions of original soils, would be less aerated and tend to retain moisture. Soil moisture can affect degradation products. Boul et al. (1994) found that increased soil moisture could enhance the loss of DDT through the creation of anaerobic microenvironments for micro-organisms able to degrade *p,p'*-DDT via *p,p'*-DDD and/or

the abiotic reductive dechlorination of *p,p'*-DDT to *p,p'*-DDD in anaerobic microenvironments.

Following release of the pesticide a range of loss mechanisms are possible. Quantifying the amount of degradation, which has occurred since closure of the site, is complicated by the variability of environmental half-lives for DDT, and lack of information on initial soil concentrations. Based on ΣDDT composition in surface soils, significant degradation has not occurred since closure of the station in 1950, compared to patterns observed in other environments. Degradation has occurred, but at a very reduced rate.

Where degradation occurred at this study site, it was influenced by soil characteristics, as in other studies. Based on the PCA analysis, mechanisms of degradation were consistent with those observed in other climates. In general, the environmental behaviour of ΣDDT in this study has been similar to other environments, but changes have occurred at a much-reduced rate. This is consistent with the cold Arctic climate controlling the fate and behaviour of ΣDDT.

B. Plants

Salix sp. and *Elymus* sp. represent a potential route of exposure for grazing animals in the vicinity of the site. The concentrations of ΣDDT detected in plants from this study indicate that residues in soil are biologically available to plant tissue, and have not become irreversibly bound to the soil.

The bioaccumulation of ΣDDT compounds was consistent with the frequent observation that plants tend to be relatively low accumulators of organic contaminants.

As reviewed by Jongbloed et al. (1996), BAFs for DDT residues in plants are generally below 1.00 and often below 0.50 (Table V-2).

Table V-2: Published DDT BAFs for plants.

Pathway	BAF	Reference
soil-rice	0.55	Verma & Pillai, 1991
soil-maize	0.16	Verma & Pillai, 1991
soil to leaves	0.05[1]	Jongbloed et al., 1996
root uptake	0.014	O'Connor Associates, 1996 in CCME, 1998a

1. Geometric mean of a review of BAFs ([kg dry soil]/[kg dry tissue]).

In this study, the BAF for *p,p'*-DDT was 0.025 for *Elymus* sp. and 0.066 for *Salix* sp. However, if an estimation of relative lipid concentration is considered, the plants exhibit almost equal BAFs. Ideally, plant pollutant concentrations should be normalized to the plant lipid concentration when directly comparing different species (Simonich & Hites, 1995).

Residue composition in plants was similar to soils. Groupings of plants in PCA reflected soil conditions. Plants containing predominantly degradation products were located on less contaminated soils and at the periphery of contaminated areas. The behaviour of plants as passive accumulators was expected. In general, lipophilic pollutants are not translocated within plants and metabolism is not significant (Simonich & Hites, 1995).

C. Mammals

The background concentration of ΣDDT in arctic ground squirrel livers in this study was 4.5 $ng \cdot g^{-1}$, for which the only compound above analytical detection limits was *p,p'*-DDE. This result was lower than published ΣDDT concentrations and consistent

with ΣDDT compositions for other species not impacted by a known point-source of contamination (Table V-3). Regional studies to determine the background load of ΣDDT in the livers of various Arctic species share two general similarities: consistently low total concentrations and *p,p'*-DDE dominated compositions.

The background concentration in this study was below concentrations reported for arctic ground squirrels (Allen-Gil et al., 1997), martens, fishers (Steeves et al., 1991), otters (Somers et al., 1987) and mink (Poole et al., 1998) (Table V-3). Compared to arctic ground squirrels, high ΣDDT concentrations in martens, fishers, otters, and mink were expected because, as predators, these animals accumulate contaminants in their tissues even at low environmental concentrations. Omnivorous arctic ground squirrels are expected to have lower contaminant concentrations in the north, but a wide range of concentrations from 5.4-1800 $ng \cdot g^{-1}$ ΣDDT have been reported for arctic ground squirrels in Northern Alaska (Allen-Gil et al., 1997). In all of the above studies, the ΣDDT composition in tissues was dominated by DDE.

The predominance of *p,p'*-DDE in livers suggests either past DDT exposure, or exposure via long-range transport. Past exposure to DDT results in the production of both DDE and DDD, however DDE is more easily stored than DDD in animal tissue (Smith, 1991). Following the use of DDT in south-central Ontario in the 1950s and 1960s, ΣDDT residues in martens contained 85% DDE, and fishers contained 70% in 1981, long after the spraying of DDT had ended (Steeves et al., 1991) (Table V-3). Exposure via long-range transport results in a predominantly *p,p'*-DDE composition because of DDE's high vapour pressure. As a result, DDE is the most mobile of ΣDDT compounds, and most subject to long-range transport. For an example of animals

impacted only by long-range transport, five caribou herds examined by Elkin and Bethke (1995) contained predominantly *p,p'*-DDE. Similarly, ΣDDT compositions in otter and mink collected from forested areas in northeastern Alberta contained only DDE, and DDD was not detectable in tissues at the analytical detection limit of 0.5 $ng \cdot g^{-1}$ (Somers et al., 1987). The predominance of DDE indicates the legacy of past DDT exposure, or exposure to only DDE via long-range transport.

The influence of contamination on arctic ground squirrel tissue concentrations appears localized to the immediate vicinity of the site. For arctic ground squirrels collected on the road between the station and camp, ΣDDT concentrations were higher than the background liver concentration, but lower than liver concentrations at either the station or camp. The intermediate ΣDDT concentrations in ground squirrel livers from the road might be attributed to long-range transport of contaminants to the north, short-range transport of contaminants from the site, or exposure as resident juveniles in contaminated areas. Exposure via long-range transport of contaminants to the north has been previously discussed, but a process of short-range transport of contamination from either the station or camp might impact ground squirrels not occupying contaminated areas. The short-range transport of organic contaminants from known point-sources in the Canadian Arctic has been reported for polychlorinated biphenyls (PCBs) (Bright et al., 1995). Exposure via short-range transport occurs as contaminated soils are aerially transported from contaminated areas and deposited on soils and vegetation up to 20 km away.

The intermediate ΣDDT concentrations in arctic ground squirrels collected on the road between the station and the camp might also have originated as exposures received

as juveniles at either the station or camp. Juvenile exposure could occur via the ingestion of contaminated food while resident at the station and camp, as modelled, or via the maternal excretion of chlorinated hydrocarbon insecticides in milk. The lipid content of milk (3-5%) and high blood flow to breast tissue can lead to considerable concentration of these chemicals compared to that in other tissues, and ingestion of maternal milk can lead to toxic effects in the juvenile recipient (Smith, 1991). Following such exposure, these juveniles could relocate. Arctic ground squirrels, especially males, disperse from the territories in which they were born (Batzli & Sobaski, 1980). Such dispersal might explain the intermediate tissue concentrations of ΣDDT in arctic ground squirrels on the road between the station and camp.

Tissue concentrations of ΣDDT decreased with distance from either the station or camp. Maximum concentration of ΣDDT in livers were 4300 $ng \cdot g^{-1}$ at the station, 1400 $ng \cdot g^{-1}$ at the camp, and 33 $ng \cdot g^{-1}$ from the road. This distinction between animal samples from the station, camp, and the road is reflected in the PCA. Two groupings occur along principal component 1 on the basis of ΣDDT concentration, with samples in the lower concentration group occurring further from either the camp or station. The tissue concentrations encountered at the station and camp are consistent with other rodents exposed to local DDT use in the Canadian Arctic. A scenario comparable to that at Kittigazuit was studied in the vicinity of Fort Churchill, Manitoba (Table V-3). Three years following aerial spraying for mosquito control, sampling within treated areas showed that the livers of collared lemmings (*Dicrostonyx groenlandicus*) contained

Table V-3: Published concentrations and compositions of ΣDDT compounds in the livers of various mammals. Samples collected from areas with and without use of DDT pesticides. Time elapsed between final application and sample collection is indicated.

Organism	Location	Time (years)	Concentration ($ng \cdot g^{-1}$)	Composition	Reference
collared lemming	Fort Churchill, Manitoba	3	5400-41000[1]	71% DDD 25% DDE 4% DDT	Brown & Brown, 1970
red squirrel	Fort Churchill, Manitoba	3	7400-17000[1]	54% DDD 43% DDE 3% DDT	Brown & Brown, 1970
collared lemming	Fort Churchill, Manitoba	NA[2]	1200[1]	48% DDD 35% DDE 17% DDT	Brown & Brown, 1970
red squirrel	Fort Churchill, Manitoba	NA[2]	1300[1]	49% DDD 32% DDE 19% DDT	Brown & Brown, 1970
arctic ground squirrel	Brooks Range, Alaska	NA	5.4-1800[3]	>85% DDE	Allen-Gil et al., 1997
martens	Algonquin, Ontario	20-30	320[4]	85% DDE 11% DDT 4% DDD	Steeves et al., 1991
fishers	Algonquin, Ontario	20-30	310[4]	70% DDE 21% DDT 9% DDD	Steeves et al., 1991
otter	northern Alberta	NA	47-70[1]	100% DDE	Somers et al., 1987
mink	western Northwest Territories	NA	23-190[5]	NA	Poole et al., 1998
arctic ground squirrel	Kittigazuit (station)	50	2.3-4300	55% DDD 42% DDE 4% DDT	Current Study
arctic ground squirrel	Kittigazuit (camp)	50	7.3-1400	63% DDD 27% DDE 10% DDT	Current Study
arctic ground squirrel	Kittigazuit (road)	NA	3.1-33	60% DDD 36% DDE 4% DDT	Current Study
arctic ground squirrel	Kittigazuit (background)	NA	4.5	100% DDE	Current Study

Note: NA indicates data either were either not available or not applicable. Refer to footnote if indicated

1. For comparative purposes, published data were lipid-corrected using the mean lipid percentage (4.3%) for livers of arctic ground squirrels collected for this study.
2. Samples collected from areas that did not receive direct DDT application, but untreated areas were approximately 3-11 km from treated areas.
3. Data was lipid-corrected based on mean lipid percentages for each of the three study populations.
4. Mean concentrations were lipid-corrected using mean lipid percentages in martens and fishers.
5. Data was lipid-corrected based on mean lipid percentages for study populations.

ΣDDT concentrations of 5400-41,000 ng·g^{-1} in the livers, with a composition of 71% DDD, 25% DDE and 4% DDT and. Liver tissues of red squirrels (*Tamiasciurus hudsonicus*) contained ΣDDT concentrations of 7,400-17,000 ng·g^{-1} with a composition of 54% DDD, 43% DDE, and 3% DDT at concentrations of in the livers (Brown & Brown, 1970).

Given the 50 years which have passed since the last DDT application at Kittigazuit, a relatively high percentage of *p,p'*-DDE was expected, but this study found that much of the ΣDDT was also *p,p'*-DDD. The large proportion of *p,p'*-DDD, 61.8%, in the study population indicated that ingested *p,p'*-DDT was being metabolized. In comparison, the composition of ΣDDT in martens and fishers 20-30 years following DDT exposure was 85% and 70% DDE, respectively. Since DDD is an intermediate metabolite of DDT, the high percentage of DDD in arctic ground squirrels in this study indicates current exposure to DDT.

BAFs in this study were calculated as either a plant tissue-animal tissue or soil-animal tissue pathway. Estimates of plant tissue-animal tissue BAFs vary significantly among studies (Table V-4). Calculated plant tissue-animal tissue BAFs in this study were low, but approached published values (Table V-4). For example the plant tissue-animal tissue BAF for meadow voles, 0.85 (Forsyth & Peterle, 1984), is close to the upper values of BAFs for *Salix* sp.-animal tissue and *Elymus* sp.-animal tissue, 0.40 and 0.98, respectively. Calculated soil-animal tissue BAFs were well below published values for this direct pathway (Table V-4). BAFs for soil-animal tissue were also low compared to published values. Similar to the plant tissue-animal tissue BAFs above, the upper range of calculated values for the soil-animal tissue BAF, 0.026, approached a published

value of 0.067 (Jongbloed et al., 1996). The BAF value of 0.067 for a soil-leaf-animal tissue food chain represents the median BAF following a review of published values for various plants and rodents.

The low BAFs determined in this study likely reflect relatively low usage of contaminated areas, and subsequent exposure, by arctic ground squirrels. Ideally, BAFs should represent the environmental concentration to which an animal is exposed, which is best approximated for animals confined to a relatively small home range for which there is a known contaminant concentration. Since arctic ground squirrels probably range over a larger area than the contaminated areas alone, lower tissue concentrations result than if the animals were restricted to contaminated areas. These tissue concentrations, combined with high, but localized soil and plant concentrations, would result in the very low BAF values calculated.

Table V-4: Published DDT BAFs from plant tissue-animal tissue and soil-animal tissue food chain for rodents.

Organism (tissue)	BAF	Reference
Plant Tissue-Animal Tissue		
meadow vole (body)	0.85	Forsyth & Peterle, 1984
rat (liver)	0.09[1]	Adams et al., 1974
Soil-Animal Tissue		
collared lemming (liver)	7.7[2]	Brown & Brown, 1970
red squirrel (liver)	6.1[2]	Brown & Brown, 1970
mammal	0.067[3]	Jongbloed et al., 1996

1. Wet or dry weight basis of tissue concentration not reported.
2. Calculated from published means for mean soil, and liver concentrations.
3. Median value reviewed data for soil-leaf-mammal food chain.

In all scenarios, the calculated TDI was below the published rodent NOEL of 0.37 $mg \cdot kg^{-1} \cdot day^{-1}$, which is a value considered to be in reasonable agreement with other

estimates of the threshold for induction of various enzymes in the rat (Smith, 1991). Considering the range of TDI estimates at the station and camp based on a diet of either *Salix* sp. or *Elymus* sp., exceeding the NOEL value was possible. Diets predominantly comprised of *Salix* sp. were most likely to exceed the NOEL. Higher TDIs from Salix sp. based diets are to be expected considering the higher BAFs for *Salix* sp., 0.066 for *p,p'*-DDT, compared to *Elymus* sp., 0.025 for *p,p'*-DDT.

In all cases the expected (i.e. calculated based on TDI values) liver burdens were well above the observed concentrations. For example at the station, the observed mean *p,p'*-DDT concentration for arctic ground squirrels was 0.18 $ng \cdot g^{-1}$, whereas the expected mean *p,p'*-DDT concentration using TDI values were 4100 $ng \cdot g^{-1}$ given a *Salix* sp. diet, and 2200 $ng \cdot g^{-1}$ given an *Elymus* sp. diet. Expected liver burdens were also calculated using a second soil-leaf-animal tissue BAF of 0.067 (Jongbloed et al., 1996). Using this value, expected *p,p'*-DDT liver burdens were again higher than the observed liver burdens, but to a lesser extent. The expected *p,p'*-DDT liver burden for arctic ground squirrels at the station was 330 $ng \cdot g^{-1}$, and the observed mean ΣDDT concentration was 74 $ng \cdot g^{-1}$. Comparison with ΣDDT, rather than *p,p'*-DDT, accounts for DDT lost via metabolism to either DDE or DDD.

The difference between the expected and observed concentrations was probably due to assumptions made during calculations. First, arctic ground squirrels were assumed to obtain their diet entirely from either the station or the camp. Considering arctic ground squirrels can move up to 1 km within a day during foraging (Batzli & Sabaski, 1980), it is possible that much of their diet was from non-contaminated areas. If so, the calculated TDI values overestimate the actual daily intake of animals at either the station and camp.

Second, depuration of ingested *p,p'*-DDT was assumed to be zero. Such an assumption would be valid in the absence of DDT metabolites, however, the large proportion of DDE, and DDD in particular, indicates that ingested *p,p'*-DDT is being metabolized. As a result, depuration is likely occurring via metabolism, and the above assumption is too conservative.

Variability within the modelling of TDI also contributes to the difference between expected and observed liver burden concentrations. Modelling exposure required certain assumptions regarding the potential routes of exposure. Regarding diet, considering only vegetation intake ignored potentially 35% of an arctic ground squirrels diet. Arctic ground squirrels are opportunistic omnivores with a dietary composition of 65-93% grasses and other plants, 6-34% seeds and berries, and 1-5% animal matter (Batzli & Sabaski, 1980). Also, there is variability among plant ingestion. Foraging patterns indicate deciduous shrubs, such as *Salix* sp., are highly palatable, whereas among grasses, *Elymus* sp. is considered among the least palatable (Batzli & Sobaski, 1980). In addition to the ingestion of contaminated plants, the only other route of exposure examined was the ingestion of contaminated soil.

For animals, inadvertent soil ingestion is an everyday occurrence related to ingestion of soiled plants, coat and muzzle licking, and inhalation of dust (Sheppard, 1998). Three specific pieces of information are needed to estimate exposure: the amount of soil ingested, the concentration of the ingested soil, and the bioavailability of the contaminant on the ingested soil (Sheppard, 1998). Exposure resulting from the ingestion of water, inhalation of contaminated air, and dermal contact was not included in the calculation of TDI. Exposure was assumed to occur indirectly via contaminated food. In

addition to contaminants incorporated into plant tissues, the amount of soil adhering to plants can be very significant. For example, 450 g of dry soil can adhere to every kg of dry annual plants (Sheppard, 1998).

Besides the ingestion of contaminated plants and soil, other routes of exposure were rejected in the calculation of TDI. Dermal contact was ignored as a route of exposure. DDT is poorly absorbed by the skin from solutions, and the absorption of solid material is so poor that it is difficult or impossible to measure either the uptake of DDT or its effect (Smith, 1991). Exposure via dermal contact and subsequent absorption was not considered to be a significant exposure pathway in this study.

Behavioural avoidance can lead to decreased exposure. Animals offered food containing high concentrations of DDT often eat little or nothing and lose weight rapidly. However, the same animals will show excellent appetites when offered the same kind of food containing little or no DDT just after refusing the major portion of their daily ration of contaminated food (Smith, 1991).

Potential exposure via the inhalation of dust particles was ignored in this study. Most DDT dust has such large particle size that any that is inhaled is deposited in the upper respiratory tract and eventually swallowed. Accordingly, toxicity data indicate that respiratory exposure to DDT is insignificant (Smith, 1991).

Though the estimated dose determined by this study was lower than the NOEL, the potential exists for these residues to become mobile from storage in lipid. Chronic exposure to DDT at low doses could produce effects when large accumulated stores are released during a starvation event such as hibernation. For ground squirrels, approximately eight to ten months are spent in hibernation, during which stored energy is

utilized (Yukon Department of Renewable Resources, 2000). Mobilisation of fat in adipose tissue due to starvation or other reasons, such as migration or hibernation, can also release stored chlorinated hydrocarbon insecticides into the circulation, sometimes with marked effects (Smith, 1991).

The mobility of ground squirrels and the localized distribution of contamination at the study site introduced the greatest uncertainty when modelling exposure. As discussed by Menzie et al. (1992), foraging over wide areas increases the variability of estimated diets. Thus, exposures will depend on factors such as the size of the contaminated area relative to the foraging area and food preference. Consequently, if foraging areas extend outside the contaminated areas, exposures are reduced due to the intake of food that is comparatively free of contamination, and results in uncertainties of several orders of magnitude (Menzie et al., 1992).

Similar to the conclusions of Menzie et al. (1992), none of the examined methods can be used directly to predict population-level effects. They offer insight into exposure and effects at the individual and perhaps brood or local levels, but such effects could eventually be manifested at the population level if severe and extensive enough. As a result, effects at the individual or local level serve as warning signs of potential population-level effects and indicate that the population is at some risk.

Despite low estimated TDI values, evidence suggest that exposure of arctic ground squirrels to *p,p'*-DDT has produced an effect. In this study, a statistically significant relationship was found between liver size and ΣDDT concentration. The relationship between liver size and ΣDDT concentrations suggests a compensatory response by arctic ground squirrels to ΣDDT exposure, which likely occurred through the

ingestion of contaminated plants or soil. Organ increase is often associated with exposure to organochlorines and correction of organ weight to body weight allows for comparison among animals at different levels of exposure. The technique has been applied to adrenal glands from voles at Love Canal, New York (Rowley et al., 1983). Calculation of LSI allows for the detection of liver increase as a compensatory response to contaminants and has been applied to arctic ground squirrels (Allen-Gil et al., 1997). It should be noted, however, that the correlation identified in this study does not necessarily demonstrate a directly causal relationship between liver size and DDT exposure. Changes in liver size also arise from infection. For example, exposure to *Escherichia coli* endotoxin can result in liver mass increase (Qian & Brosnan, 1996).

Exposure to DDT has been demonstrated to be an inducer of microsomal mixed function oxygenase enzymes of the liver, and mild to severe hepatic effects in experimental animals have resulted from acute, sub-chronic, and chronic oral administration of DDT (ASTDR, 1994). As reviewed by Smith (1991), the evidence is strong that enlargement of the liver and individual liver cells is adaptive at dosages where the increase in endoplasmic reticulum is accompanied by a parallel increase in the activity of enzymes of the cytochrome P-450 system. Return of the liver to normal size also occurs if dosage is discontinued soon enough (Kunz et al., 1966 in Smith, 1991). Since effects are reversible in early stages, the livers of arctic ground squirrels with lower ΣDDT concentrations may have returned to normal.

Contamination at the station and camp was responsible for the observed ΣDDT concentrations in the livers of arctic ground squirrels. Arctic ground squirrels from background and road locations contained ΣDDT concentrations and compositions

suggesting they were not impacted by contamination at the station or camp, whereas liver concentrations and compositions at the station and camp were consistent with a recently sprayed area. The composition of ΣDDT in the livers of arctic ground squirrels indicates that contamination at the station and camp was not representative of a typical legacy situation. The predominance of DDD indicates current metabolism of DDT, as opposed to the predominance of DDE, which would indicate long-term storage. Estimated exposures from the station and camp are below the NOEL. However, a measurable effect appears to be occurring. Increases in liver size were significantly related to contaminant concentrations in liver tissue. Disparity between the low estimated dose and observed effect was probably due to model uncertainty.

The observed concentration and composition of ΣDDT in the livers of ground squirrels in this study was supportive of three conclusions. First, that tissues concentrations of ΣDDT were caused by contamination at the station and camp, and not from long-range transport. Second, the pattern is inconsistent with one expected for a legacy issue. Third, the dosage is sufficient to induce biotransformation via metabolic pathways.

D. Conclusions

Recently, Environment Canada identified a data gap and stated that, "data on soil residual levels [of DDT] in Canada, particularly in northern Canada, are scarce if not non-existent" (EC, 1998a). The results of this study help address that data gap. This study examined the environmental behaviour and fate of DDT pesticide in soils, sediments, plants, and animals in a terrestrial arctic environment. The results of this study indicate that the behaviour and fate of DDT in an arctic environment is different

than in temperate and tropical ecosystems; the ultimate influence of the arctic climate controlled the rate of DDT degradation. Despite the passage of 50 years since closure of the LORAN site, DDT contamination has persisted in a manner comparable to the passage of three years in a sub-arctic environment (Brown & Brown, 1970).

In soils, ΣDDT contamination has remained concentrated, localized, and non-degraded. In tropical and temperate environments, volatilisation and microbial degradation would normally account for significant DDT loss. In this study, the role of volatilisation and microbial degradation was greatly reduce due to the climate, likely temperature. The composition of ΣDDT in soils indicated that little degradation had occurred. Locations where DDT had degraded to either DDE or DDD were consistent with behaviour in temperate climates. The composition of ΣDDT in sediments indicated that DDT degradation comparable to temperate ecosystems could occur, once the influence of temperature was reduced. The ΣDDT contained in soils presented a source of contaminants into the terrestrial food chain.

Concentrations of ΣDDT in plants demonstrated the mobility of contaminants from the soil, and plants were a potential route of contaminant exposure for terrestrial animals. The ΣDDT composition in *Salix* sp. and *Elymus* sp. demonstrated organochlorine accumulation, without subsequent metabolism once in plant tissue. Consistent with published data, soil-plant tissue BAFs indicated the uptake of contaminants by plants was low. Different BAFs between Salix sp. and Elymus sp. were the likely the result of different lipid contents between the plants.

Data for arctic ground squirrels provided a mammalian receptor with which to investigate the impact of DDT contamination. Arctic ground squirrels from areas not

impacted by contamination at the station and camp contained ΣDDT lower than other terrestrial mammals in remote locations in Canada. Arctic ground squirrels in close proximity to either the station or camp accumulated ΣDDT concentrations comparable to those from a recently sprayed area in a sub-arctic environment (Brown & Brown, 1970).

The composition of ΣDDT compounds in animal tissue indicated the persistent hazard posed by contamination at the site. Tissue compositions were not indicative of historical exposure as expected, but rather current exposure. The intermediate degradation product, DDD, as opposed to the long-term storage product, DDE, dominated compositions. The known ecology of arctic ground squirrels allowed for the modelling of DDT exposure through the ingestion of contaminated plants and soil. Despite the low estimated dosage, a toxicological effect was observed in arctic ground squirrels. Increases in liver size were significantly correlated with ΣDDT concentrations. The life cycle of arctic ground squirrels might explain the observed effect. During hibernation, arctic ground squirrels rely on stored fat, and during metabolism, potentially toxic amounts of ΣDDT could be released and result in the observed effects.

The DDT contamination at Kittigazuit provided a unique opportunity to examine the environmental behaviour and fate of DDT pesticide in soils, sediments, plants, and animals in a terrestrial arctic environment. A combination of characteristics separates this study from others. This study is unique because it deals with a known point-source of DDT contamination, located in an arctic ecosystem, and with a known history of localized DDT use.

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APPENDIX A: MAPS OF SAMPLE LOCATIONS

Appendix A1: Soil Sample Locations at Station

Appendix A2: Soil Sample Locations at Station (South)

Appendix A3: Soil Sample Locations at Camp

Appendix A4: Plant Sample Locations at Station (South)

Appendix A5: Plant Sample Locations at Camp

Appendix A6: Animal Sample Locations at Station (North)

Appendix A7: Animal Sample Locations at Station (East)

Appendix A8: Animal Sample Locations at Station (North)

Appendix A9: Animal Sample Locations at Station (South)

Appendix A10: Animal Sample Locations at Road

Appendix A11: Animal Sample Locations at Camp

Appendix A1: Soil Sample Locations at Station

13273
13270
13017
13269
13272
13023
Steel Debris
13271
13016
13068
13061
13203
13067
13062
Building Foundation
13063
13202
13201
13010
Debris From Wall
13066
Steel Pipes
13204
13009
13012
13064
Wood Debris
13008
Boiler House Foundation
Steel Debris
13007
13006
13013-4
13065
Battery
13074
13200
13015
13073
13199
Building Foundation
Boiler House
13076
10207
13198
13004
13003
13071-2
13075
13005
13001-2
13263
13261
13082
13083-4
13246
Steel Post
Road
13245
13077
Steel Pilings
13081
13078
13080
13079
Warehouse Pad
13196
13197
10273
10193
10195
10194
10196
10186
10188
10187
10272
13097
10197
10201
13096
13098-9
13100
Garage Foundation
13095
13092
13195
13193-4
13094
13093
10202
N
W
E
S
Metres
0
6
12
Pole
13192
13186
Pole

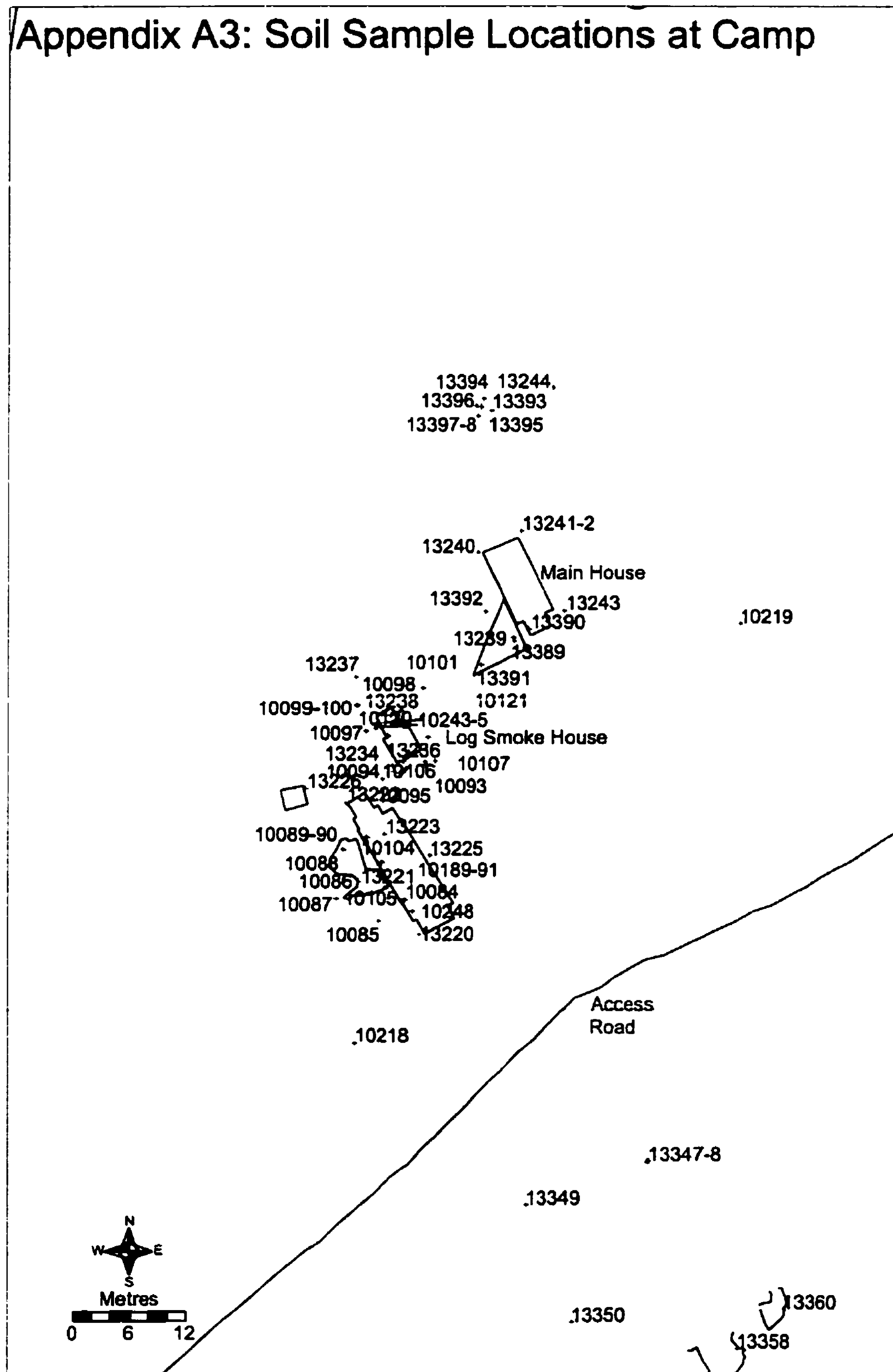
Appendix A2: Soil Sample Locations at Station (South)

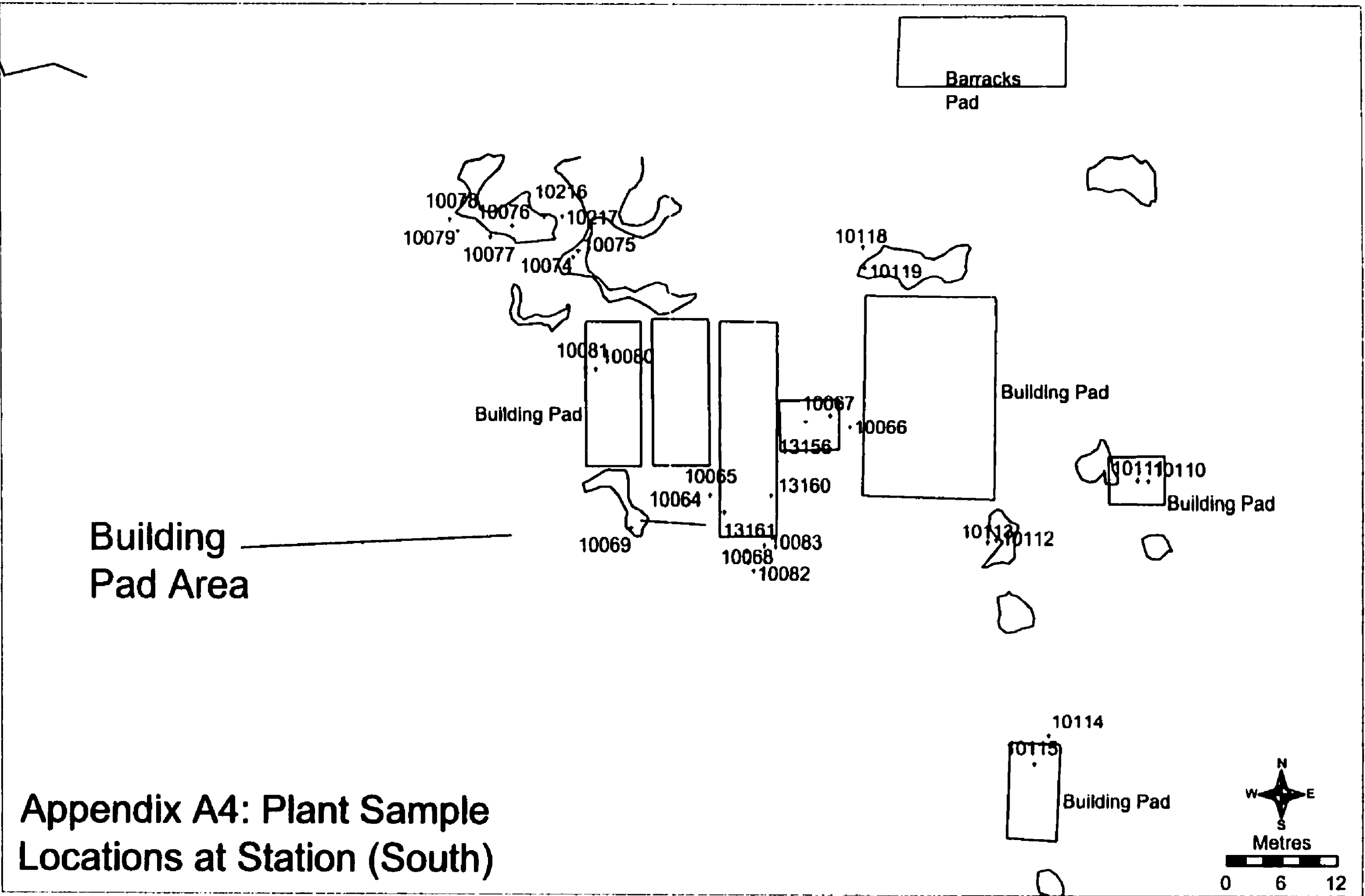
Building Pad Area

N
W E
S

Metres

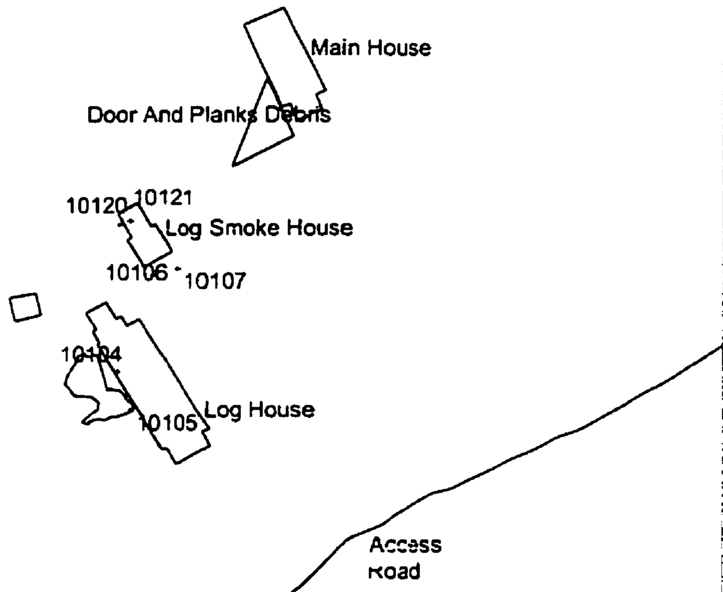
0 6 12

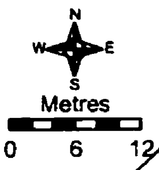

Appendix A3: Soil Sample Locations at Camp
13394 13244
13396 13393
13397-8 13395
13241-2
13240
Main House
13392
13243
13390
10219
13389
13237
10101
13391
10098
10121
10099-100
13238
10243-5
10097
Log Smoke House
13234
10107
10094
10093
10089-90
13223
10104
13225
10189-91
10088
10086
13221
10087
10105
10084
10085
13220
Access Road
10218
13347-8
13349
13350
13360
13358
N
W
E
S
Metres
0
6
12

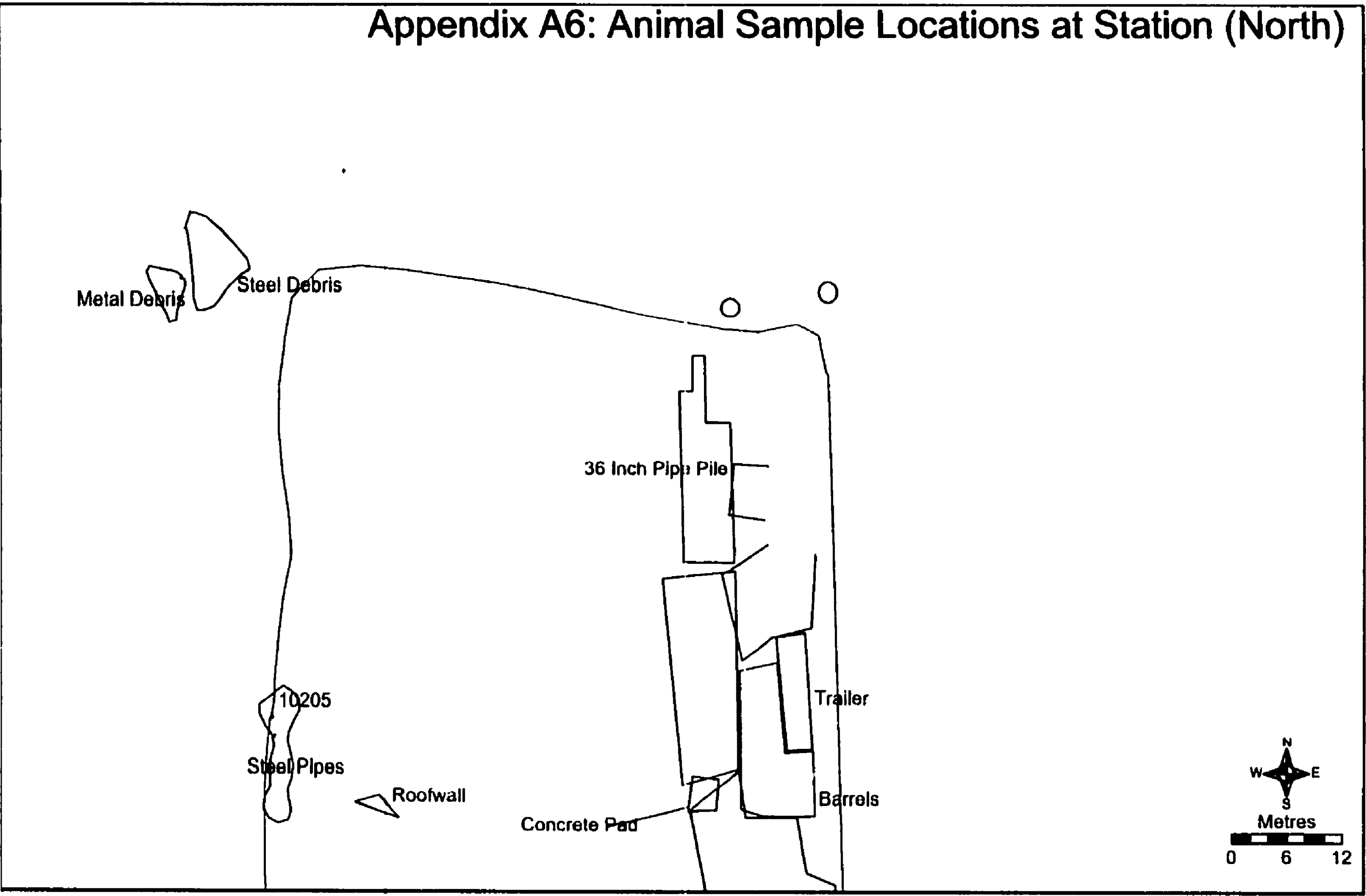


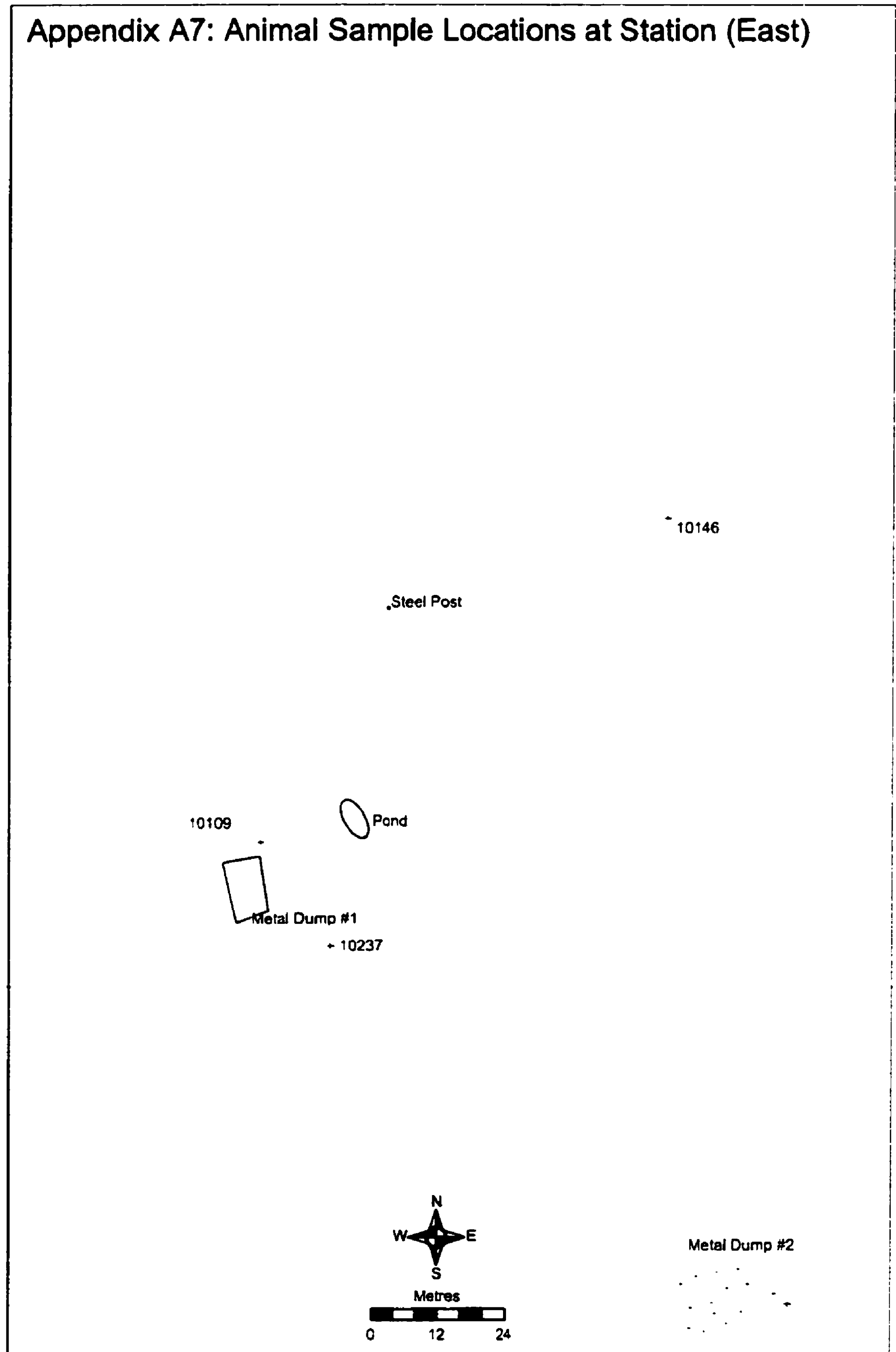
Appendix A4: Plant Sample Locations at Station (South)

Appendix A5: Plant Sample Locations at Camp

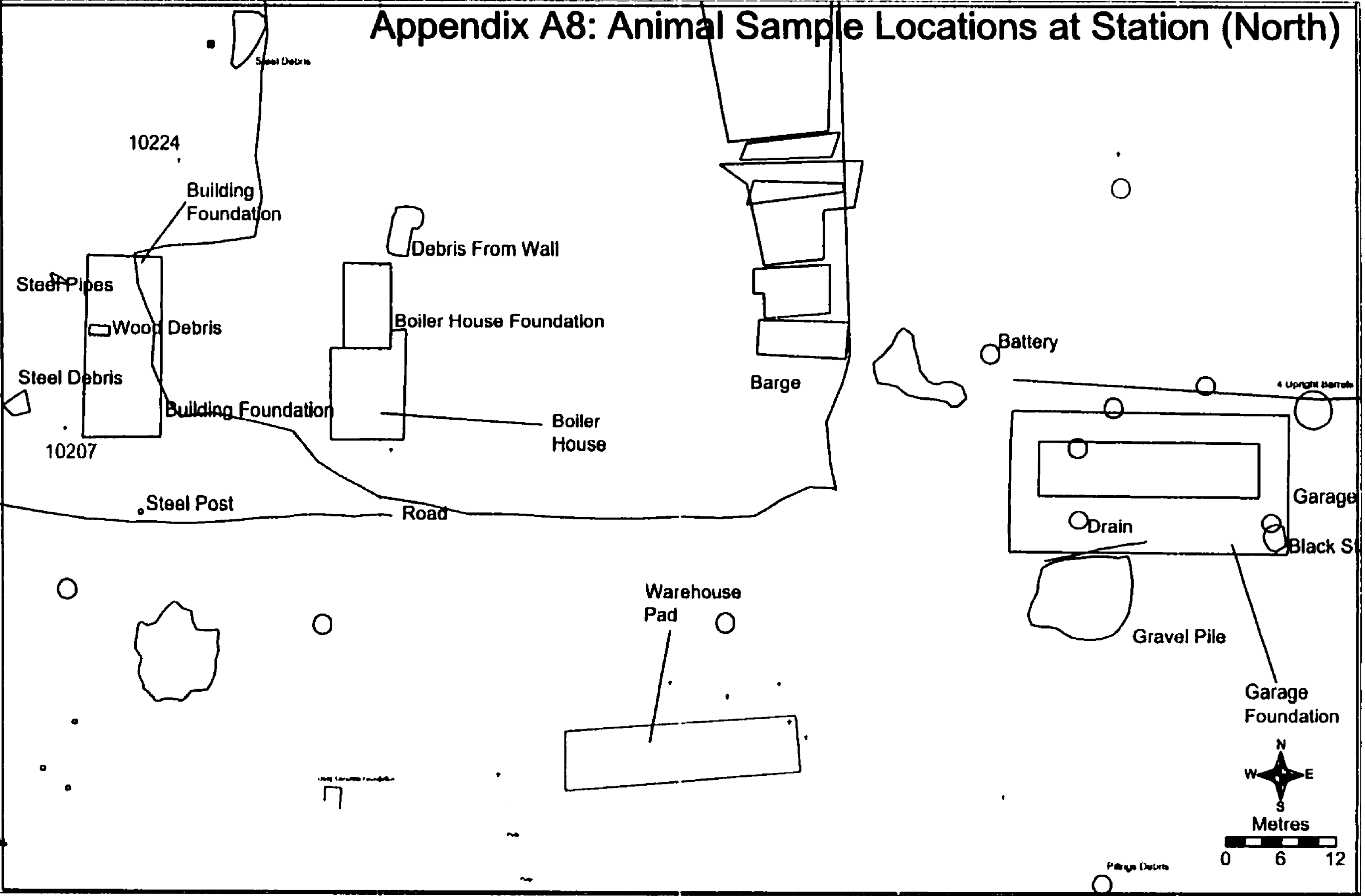


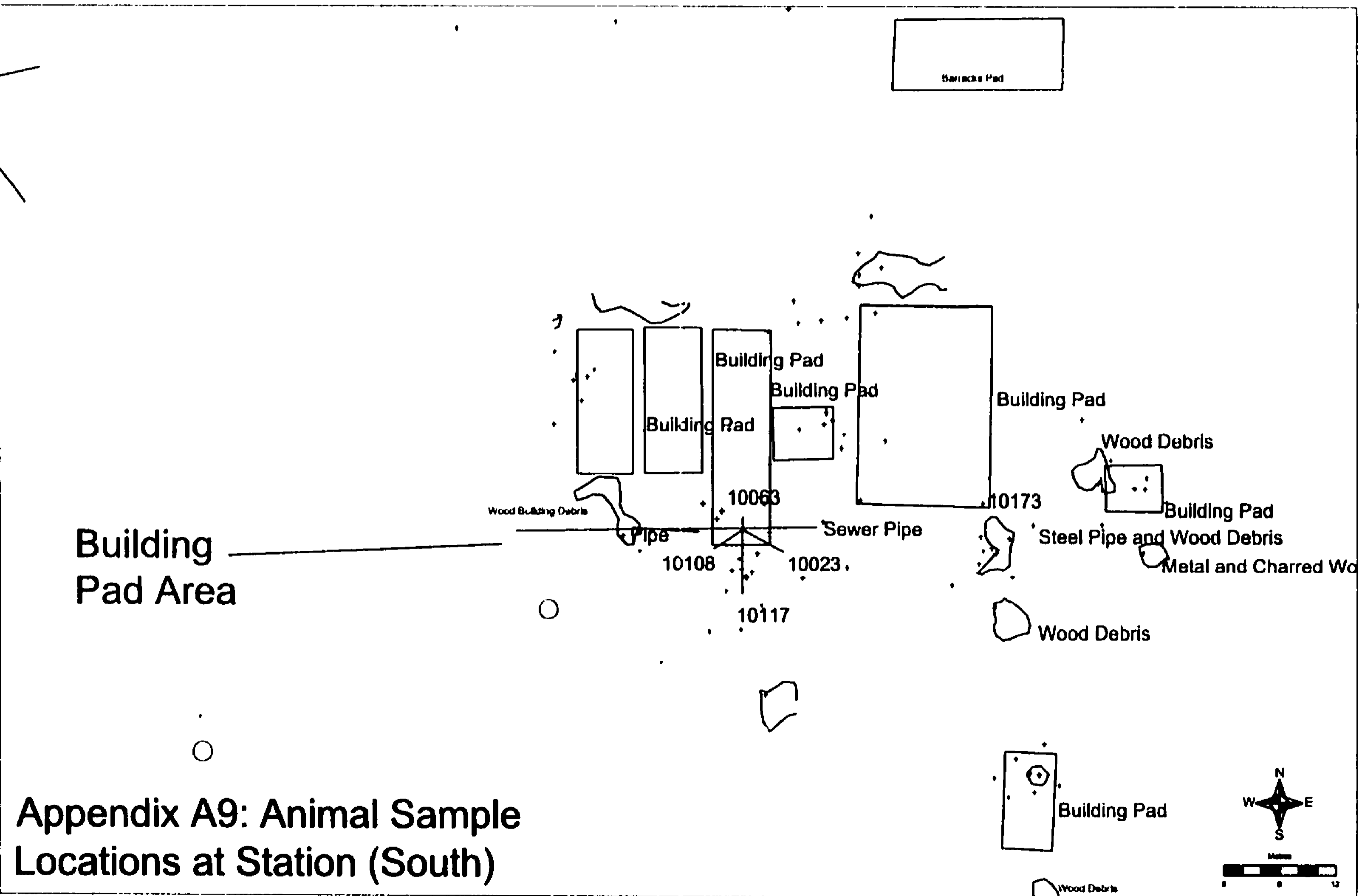



Appendix A6: Animal Sample Locations at Station (North)
Metal Debris
Steel Debris
36 Inch Pipe Pile
10205
Steel Pipes
Roofwall
Concrete Pad
Trailer
Barrels
N
W
E
S
Metres
0
6
12

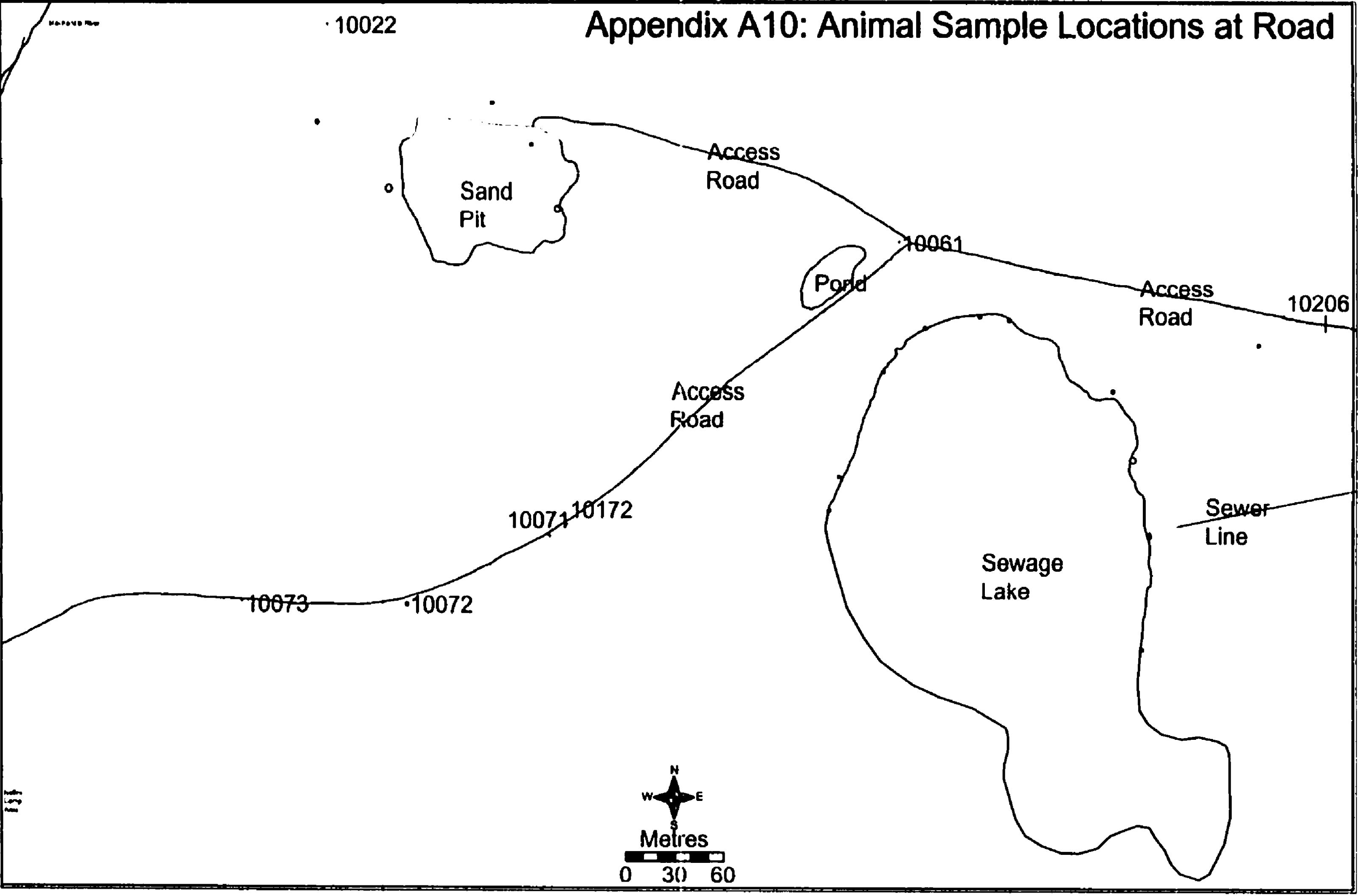

Appendix A7: Animal Sample Locations at Station (East)
10146
Steel Post
10109
Pond
Metal Dump #1
10237
N
W
E
S
Metres
0
12
24
Metal Dump #2

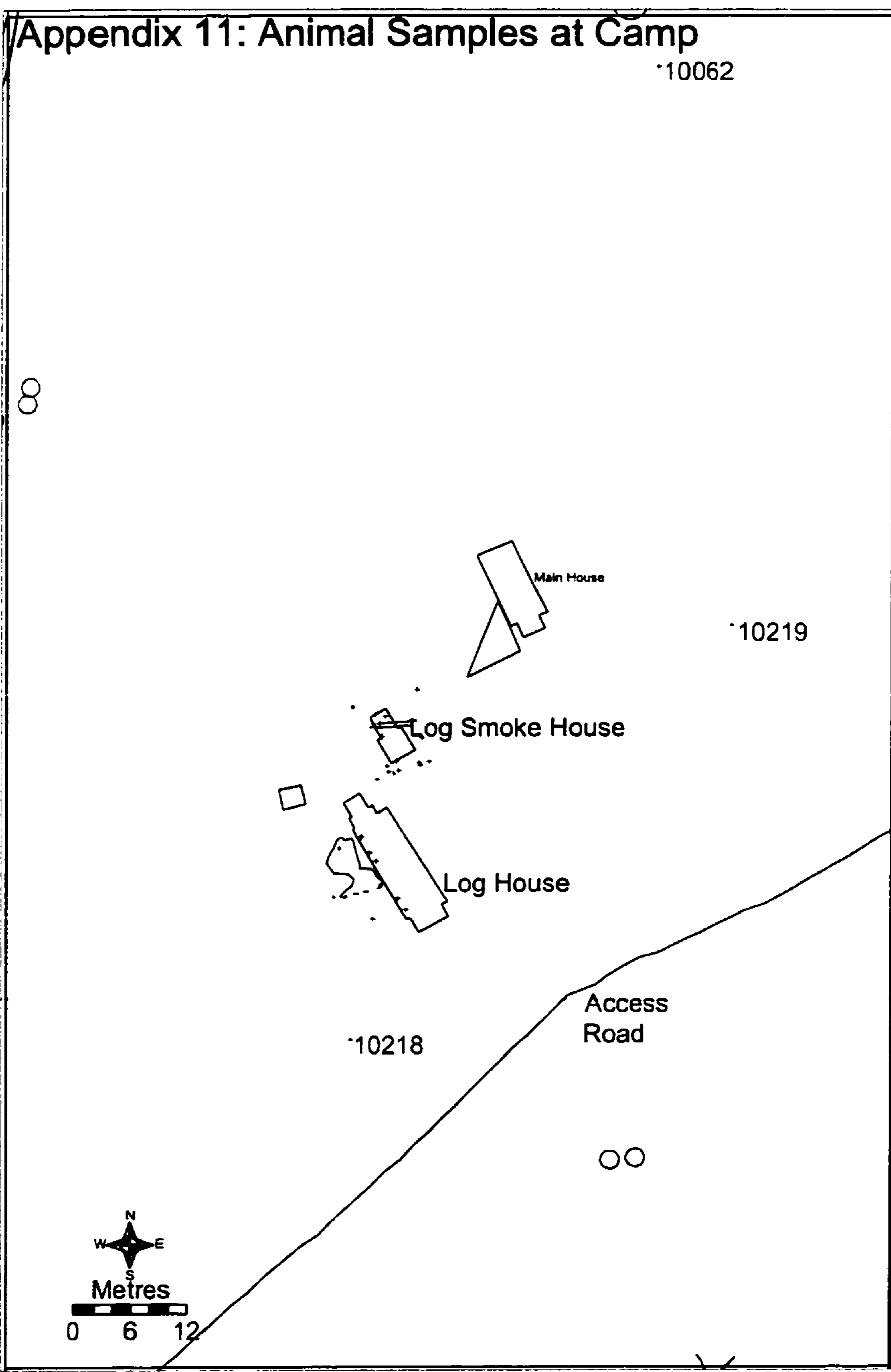
Appendix A8: Animal Sample Locations at Station (North)





Appendix A9: Animal Sample Locations at Station (South)


Appendix A10: Animal Sample Locations at Road
10022
Sand Pit
Access Road
10061
Pond
Access Road
10206
Access Road
10071
10172
10073
10072
Sewage Lake
Sewer Line
N
W
E
S
Metres
0 30 60


Appendix 11: Animal Samples at Camp
10062
Main House
10219
Log Smoke House
Log House
Access
Road
10218
N
W
E
S
Metres
0 6 12

APPENDIX B: DATA

Appendix B1: Analytical Data for Soils and Sediments

Appendix B2: Analytical Data for Plants

Appendix B3: Analytical Data for Mammals

Appendix B1: Analytical Data for Soils and Sediments.

		UTM Coordinates		Compound (ng/g)					
Sample	Matrix	Eastings	Northings	o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	o,p'-DDT	p,p'-DDT
10001	soil	544288.01	7686041.68		10	10	10	10	10
10004	soil	544301.84	7686040.14		12593	3950	8058	45192	137667
10006	soil	544310.53	7686031.54		24	34	97	101	478
10009	soil	544341.00	7686016.15		131	96	257	167	696
10010	soil	544341.00	7686016.16		160	106	269	134	733
10011	soil	544338.32	7686009.86		5478	1868	4944	14514	146541
10013	soil	544328.47	7686006.47	15	98	828	2198	792	2505
10019	soil	544334.63	7686019.13		169	69	147	430	1312
10020	soil	544334.61	7686019.14		184	74	178	439	1541
10032	soil	544352.90	7685998.61		10	10	40	10	243
10036	soil	544369.26	7686012.24		28	10	10	10	116
10038	soil	544368.84	7686004.42		158	25	144	186	1431
10047	soil	544358.22	7685984.15		10	64	168	10	106
10051	soil	544338.19	7686032.52		211	107	301	668	2735
10052	soil	544334.14	7686032.30	28	499	1290	1984	1689	5256
10059	soil	544305.18	7686017.94		313	48	96	630	1341
10060	soil	544305.12	7686017.94		303	47	99	599	1389
10085	soil	543152.43	7685934.99		44	10	10	10	100
10087	soil	543148.01	7685937.42		30	10	10	10	69
10088	soil	543148.67	7685942.78		146	10	53	53	344
10092	soil	543157.41	7685952.25		33	10	30	10	78
10095	soil	543152.86	7685950.43		699	258	1224	3652	64783
10099	soil	543150.14	7685958.38		253	10	78	147	1067
10100	soil	543150.14	7685958.49		191	10	53	91	614
10101	soil	543157.17	7685960.31		10	10	10	10	10
10102	soil	543157.70	7685954.97	38	5011	542	2554	6537	41875
10124	soil	544312.60	7686037.06		334	77	280	692	4109
10126	soil	544313.86	7686031.24		320	230	402	632	2048
10129	soil	544334.05	7686029.09		244	68	153	840	2167
10130	soil	544334.03	7686029.02		271	62	167	659	2344
10131	soil	544331.57	7686028.75		593	273	1641	954	8209
10133	soil	544329.91	7686033.90		27	10	10	10	92
10135	soil	544336.91	7686002.75		162	89	259	845	6922
10139	soil	544321.96	7685995.93		10	10	37	21	270
10140	soil	544321.94	7685995.92		10	10	30	10	233
10143	soil	544333.47	7685988.32		2971	348	1880	6838	56604
10145	soil	544342.54	7685987.53		23	10	38	28	176
10148	soil	544518.33	7685845.08		10	10	10	10	10
10194	soil	544287.19	7686082.45		68	10	40	36	338
10196	soil	544297.62	7686081.91		68	45	77	64	176
10204	soil	544311.90	7686059.90		75	10	83	10	377
10212	soil	544302.39	7686044.69		2128	106	525	1322	18804
10214	soil	544292.75	7686051.91		10	10	10	10	10
10230	sed.			0.08	0.4	0.37	1.2	0.0015	0.01
10232	sed.			0.004	0.18	0.08	0.39	0.04	0.17
10234	sed.			0.0025	0.0035	0.004	0.02	0.002	0.006
10236	sed.			0.0035	0.0045	0.004	0.011	0.0015	0.002
13001	soil	544250.32	7686108.84		120	10	59	158	991
13002	soil	544250.30	7686108.86		139	10	59	172	1076
13003	soil	544247.28	7686109.38		10	10	10	10	10

Appendix B1: Analytical Data for Soils and Sediments.

Sample	Matrix	UTM Coordinates		Compound (ng/g)					
		Eastings	Northings	o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	o,p'-DDT	p,p'-DDT
13006	soil	544243.18	7686116.39		10	10	10	10	30
13015	soil	544251.32	7686114.12		29	10	10	10	161
13026	soil	544227.30	7686329.91		10	10	10	10	10
13036	soil	544250.70	7686392.81		10	10	10	10	10
13056	soil	544290.85	7686168.10		10	10	10	10	10
13066	soil	544289.18	7686127.08		10	10	10	10	10
13071	soil	544318.38	7686112.05		45	10	20	37	156
13072	soil	544318.41	7686112.03		49	10	10	41	171
13076	soil	544350.33	7686113.96		10	10	10	10	10
13077	soil	544351.11	7686101.89		10	10	10	10	10
13080	soil	544335.30	7686096.46		56	108	251	194	772
13081	soil	544315.88	7686098.23		233	10	108	65	855
13082	soil	544326.66	7686106.85		10	10	10	10	32
13085	soil	544320.41	7686038.98		108	27	88	104	674
13087	soil	544324.39	7686038.68		10	31	113	29	177
13088	soil	544314.87	7686049.27		28	10	40	30	234
13090	soil	544311.17	7686043.76		10	10	10	10	24
13091	soil	544308.56	7686049.05		58	10	78	29	471
13092	soil	544295.85	7686075.11		10	10	10	10	10
13095	soil	544268.00	7686072.92		10	10	10	10	25
13098	soil	544280.68	7686074.86		10	10	10	20	56
13100	soil	544294.07	7686079.45		908	562	1024	1532	4658
13101	soil	544361.90	7686054.41		10	10	10	10	10
13104	soil	544350.77	7686059.83		10	10	10	10	10
13105	soil	544343.83	7686060.18		10	10	32	30	78
13106	soil	544343.53	7686055.77		10	10	10	10	10
13109	soil	544350.21	7686057.46		10	10	10	10	10
13111	soil	544345.38	7686041.85		10	44	104	10	22
13112	soil	544343.23	7686033.62		132	42	89	271	765
13113	soil	544336.89	7686035.15		246	106	282	410	1991
13115	soil	544362.42	7686023.86		36	23	55	25	82
13117	soil	544366.74	7686009.44		336	78	253	445	2011
13118	soil	544359.61	7686012.80		10	31	67	10	95
13119	soil	544359.62	7686012.81		10	30	62	10	84
13120	soil	544352.70	7686003.91		317	302	698	419	2474
13121	soil	544345.59	7685999.01		115	86	242	123	745
13122	soil	544348.84	7685989.71		42	26	71	71	220
13124	soil	544357.09	7685980.70		42	93	213	522	1920
13125	soil	544355.67	7685976.84		10	10	10	10	10
13126	soil	544351.34	7685977.94		146	21	46	141	387
13127	soil	544351.38	7685977.89		320	48	86	321	1054
13128	soil	544355.69	7685973.43		10	10	10	10	10
13129	soil	544356.67	7685967.89		20	10	10	10	69
13130	soil	544350.74	7685967.99		76	10	37	58	207
13133	soil	544367.81	7685935.15		10	10	10	10	10
13137	soil	544370.36	7685914.52		10	10	10	10	10
13138	soil	544370.29	7685914.59		10	10	10	10	10
13141	soil	544434.67	7685919.95		10	10	24	10	10
13142	soil	544433.49	7685923.39		10	10	10	10	10
13144	soil	544403.69	7685926.31		10	10	10	10	10

Appendix B1: Analytical Data for Soils and Sediments.

		UTM Coordinates		Compound (ng/g)					
Sample	Matrix	Eastings	Northings	o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	o,p'-DDT	p,p'-DDT
13148	soil	544353.81	7685954.31		10	10	10	10	22
13151	soil	544367.78	7686041.59		48	36	89	102	324
13152	soil	544367.98	7686044.84		67	10	32	34	98
13155	soil	544339.86	7686041.68		10	38	106	10	37
13156	soil	544331.72	7686017.42		56	69	164	266	1009
13157	soil	544324.64	7686026.65		10	10	10	10	10
13158	soil	544325.42	7686018.99		10	10	10	10	10
13159	soil	544323.05	7686015.04		10	10	10	10	10
13160	soil	544327.91	7686009.59		37	10	88	41	251
13161	soil	544325.21	7686003.70		774	312	507	626	3043
13162	soil	544315.57	7686006.70		10	10	10	10	49
13163	soil	544316.44	7686013.03		24	10	32	30	102
13164	soil	544317.55	7686023.13		10	10	10	10	10
13166	soil	544314.25	7686023.55		32	10	10	49	110
13167	soil	544321.30	7686029.98		10	10	10	10	10
13168	soil	544315.52	7686028.55		170	72	215	228	1182
13170	soil	544310.88	7686025.02		39	25	79	38	192
13171	soil	544307.10	7686022.19		1009	172	452	1348	4871
13172	soil	544309.83	7686016.06		38	58	118	10	34
13173	soil	544311.75	7686009.83		10	10	10	10	10
13174	soil	544307.74	7686006.39		10	10	10	10	52
13176	soil	544303.18	7685997.62		141	128	417	130	722
13177	soil	544305.36	7686035.15		928	214	526	2274	11625
13178	soil	544298.61	7686028.05		41	10	34	26	144
13179	soil	544300.02	7686037.63		2194	465	739	2445	14448
13180	soil	544292.51	7686038.54		492	157	443	388	2373
13181	soil	544274.45	7686041.28		61	10	43	59	233
13183	soil	544253.45	7686036.75		10	10	10	10	10
13191	soil	544246.44	7686061.58		10	10	10	10	10
13198	soil	544225.93	7686109.76		10	10	10	10	10
13202	soil	544211.00	7686128.95		10	10	10	10	10
13220	soil	543156.79	7685933.54		59	10	71	65	336
13221	soil	543152.51	7685941.08		425	53	126	331	1533
13222	soil	543148.94	7685947.83		76	10	59	31	339
13223	soil	543153.08	7685944.45		77	54	169	236	1073
13225	soil	543157.94	7685942.13		30	10	98	20	91
13227	soil				10	10	10	10	10
13229	soil				10	10	10	10	10
13234	soil	543155.05	7685952.41		1610	168	710	1225	11489
13235	soil	543152.09	7685955.94		300	115	374	277	1263
13237	soil	543150.00	7685961.55		77	10	31	20	136
13240	soil	543163.06	7685975.12		38	10	24	10	105
13243	soil	543172.15	7685968.74		22	10	36	10	10
13262	soil	544250.98	7686107.81		10	10	10	10	10
13263	soil	544249.22	7686107.65		10	10	10	10	10
13318	soil	544371.41	7686043.40		25	54	145	346	1605
13323	soil	544335.89	7686019.04		280	100	492	341	1506
13324	soil	544336.04	7686015.01		565	201	1009	1207	5661
13325	soil	544329.64	7686018.41		29	28	107	108	430
13326	soil	544329.79	7686013.20		157	537	1771	385	1696

Appendix B1: Analytical Data for Soils and Sediments.

		UTM Coordinates		Compound (ng/g)					
Sample	Matrix	Eastings	Northings	o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p' DDD	o,p'-DDT	p,p'-DDT
13327	soil	544334.13	7686016.69		514	279	590	1653	6388
13333	soil	544326.20	7686010.47		10	10	10	10	10
13334	soil	544324.17	7686001.68		2326	532	817	1750	9882
13335	soil	544325.98	7686001.60		1901	507	1019	2647	18668
13336	soil	544325.32	7686006.39		497	497	1477	1434	7495
13338	soil	544315.65	7686026.52		10	10	10	10	10
13363	soil	544160.40	7685780.20		10	10	10	10	10
13367	soil	544148.37	7685782.78		10	10	10	10	48
13392	soil	543163.88	7685968.66		10	10	26	10	35
13399	soil	544434.05	7685924.52		10	10	10	10	10
13417	soil	544336.97	7686013.26		616	112	484	645	2760

Appendix B2: Analytical Data for Plants

Sample	Matrix	UTM Coordinates		Compound (ng/g)					
		Eastings	Northings	o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	o,p'-DDT	p,p'-DDT
10064	Elymus sp.	544322.74	7686007.90	0.24	22	4.4	13	43	210
10065	Elymus sp.	544321.28	7686009.51	0.005	0.4	0.02	0.1	0.3	5
10066	Elymus sp.	544336.56	7686016.82	0.69	39	4	10	29	170
10067	Salix sp.	544334.38	7686017.97	0.87	28	27	66	53	230
10068	Salix sp.	544325.42	7686002.56	1.2	170	23	57	43	230
10069	Salix sp.	544312.60	7686006.11	0.105	1.4	0.115	0.13	0.37	3
10074	Salix sp.	544306.32	7686034.79	0.51	39	2	7.5	25	200
10075	Elymus sp.	544306.86	7686035.47	0.24	28	2.5	6.6	19	140
10076	Salix sp.	544299.65	7686038.13	7.5	390	33	130	325	1550
10077	Elymus sp.	544297.30	7686036.96	0.06	14	0.3	1.1	4.7	28
10078	Salix sp.	544292.87	7686038.83	0.28	22	0.825	2	8.3	47.5
10079	Elymus sp.	544293.45	7686037.66	0.03	2.6	0.07	0.41	1.2	9.3
10080	Salix sp.	544308.77	7686022.92	0.4	42	0.9	1.7	23	81
10081	Elymus sp.	544307.54	7686023.08	0.6	17	0.8	1.2	15	45
10082	Salix sp.	544325.88	7686001.74	1.8	200	21	58	73	410
10083	Elymus sp.	544327.14	7686004.21	1	65	13	27	51	230
10104	Salix sp.	543152.07	7685942.32	0.25	7.9	0.2	0.8	6.2	26
10105	Elymus sp.	543153.19	7685938.69	0.3	10	0.25	0.6	5.7	21
10106	Salix sp.	543155.24	7685951.40	0.07	30.5	0.55	3.45	2.65	48.5
10107	Elymus sp.	543157.51	7685951.99	0.035	5.7	0.2	1.2	1.6	17
10110	Salix sp.	544369.09	7686011.12	1.45	18	5	6	9.1	19
10111	Elymus sp.	544367.94	7686011.15	0.11	0.71	0.04	0.045	0.11	1.2
10112	Salix sp.	544352.52	7686004.87	0.36	2.9	0.14	0.155	0.78	5.9
10113	Elymus sp.	544351.58	7686004.64	0.07	0.27	0.385	1.1	0.95	4.8
10114	Salix sp.	544357.75	7685980.94	0.75	1.9	0.58	2.2	9.7	41
10115	Elymus sp.	544356.69	7685981.10	0.085	0.26	0.04	0.045	0.13	0.26
10118	Salix sp.	544338.09	7686035.83	0.47	18	1.7	5.9	31	150
10119	Elymus sp.	544338.20	7686033.68	0.56	24	10	18	19	70
10120	Salix sp.	543152.21	7685956.17	0.3	13	0.9	2.8	3.4	23
10121	Elymus sp.	543153.19	7685956.55	0.25	1.8	0.1	0.3	0.9	6.4
10216	Salix sp.	544303.22	7686039.17	13	700	55	130	710	2200
10217	Elymus sp.	544305.15	7686039.15	0.245	23	1.3	3.8	17	85
10249	Salix sp.	544095.56	7685613.36	0.55	0.5	0.25	0.25	0.3	0.325
10250	Elymus sp.	544092.86	7685613.49	1.45	1.3	1.5	1.55	2.75	3
10254	Salix sp.	544765.80	7685752.07	0.3	0.3	0.35	0.35	0.45	0.5
10255	Elymus sp.	544764.89	7685753.87	0.65	0.6	0.4	0.45	0.8	0.9
13156	Salix sp.	544331.72	7686017.42	0.33	4.3	11	21	13	58
13160	Salix sp.	544327.91	7686009.59	2	55	12	54	180	1000
13161	Salix sp.	544325.21	7686003.70	0.24	31.5	4.1	14	18	105
13227	Salix sp.			0.005	0.057	0.003	0.01	0.025	0.14

Appendix B3: Analytical Data for Mammals

Sample	Matrix	UTM Coordinates		Sex (M/F)	Body Mass (kg)	Liver Mass (g wet)	Body Length (cm)	Lipid (%)	Compound (ng/g)					
		Eastings	Northings						o,p'-DDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	o,p'-DDT	p,p'-DDT
10022	S. parryi	543406.41	7686313.95					4.2	0.002	0.04	0.001	0.08	0.004	0.0025
10023	S. parryi	544325.55	7686006.80	M	0.91	55.6	30	5.2	0.0024	46	0.052	52	0.71	0.29
10061	S. parryi	543755.92	7686185.93	M	0.91	34.8	30	5	0.00285	0.12	0.003	0.16	0.0089	0.006
10062	S. parryi	543182.69	7686028.87	M	0.34	16.8	23	3.9	0.00315	5.4	0.04	11	0.11	0.055
10063	S. parryi	544323.25	7686008.80	F	0.23	14.8	21	3.1	0.00265	12	0.018	33	0.53	0.034
10071	S. parryi	543541.62	7686009.96	M	0.45	14.2	24	3.3	0.0025	0.24	0.0015	0.15	0.0025	0.0035
10072	S. parryi	543454.18	7685969.97	F	0.41	18.9	23	3.6	0.0027	0.11	0.0028	0.53	0.039	0.006
10073	S. parryi	543353.80	7685971.85	M	0.45	21.8	24	3.1	0.002	0.11	0.0015	0.2	0.006	0.0035
10108	S. parryi	544323.31	7686008.70	M	0.68	48.4	28	4.5	0.00285	42	0.056	20	0.7	0.47
10109	S. parryi	544437.32	7685929.23	M	0.79	38.4	30	5.15	0.003425	0.0355	0.002275	0.057	0.0078	0.011
10117	S. parryi	544323.26	7686008.82	M	0.57	56.6	29	7.1	0.0034	5.5	0.0071	4.4	0.12	0.047
10146	S. parryi	544510.11	7685988.67	F	0.91	42.3	28	4.9	0.00345	0.054	0.00175	0.054	0.0061	0.00395
10172	S. parryi	543552.14	7686016.66	F	0.34	13.7	23	3.6	0.09	0.12	0.05	0.06	0.12	0.16
10173	S. parryi	544351.54	7686009.59	M	0.23	15	20	3.5	0.045	52	1	97	0.035	0.05
10205	S. parryi	544237.40	7686174.54	F	0.79	37.8	30	5.4	0.0375	0.275	0.0175	0.865	0.03	0.04
10206	S. parryi	543990.66	7686142.09	F	0.68	31.5	28	4.5	0.04	0.38	0.02	0.94	0.035	0.05
10207	S. parryi	544214.65	7686111.01	F	0.57	31.4	27	4	0.003	0.8	0.003	1.6	0.02	0.006
10218	S. parryi	543149.92	7685921.75	F	0.23	14.2	21	3.8	0.15	11	0.065	42	0.125	0.17
10219	S. parryi	543190.70	7685967.39	M	0.68	31	25	5	0.045	0.06	0.025	0.13	0.045	0.06
10224	S. parryi	544226.82	7686139.09	M	0.68	28.2	29	4.8	0.08	23	0.05	58	0.125	0.17
10237	S. parryi	544449.76	7685910.20	F	0.57	49.9	28	4	0.003	0.325	0.00125	0.17	0.006	0.00375
10238	S. parryi	544091.71	7685614.69	F	0.34	18.9	24	3.8	0.055	0.17	0.045	0.06	0.105	0.14
10239	S. parryi	544091.63	7685614.55	F	0.23	15.2	21	3.6	0.065	0.09	0.03	0.04	0.075	0.1