

APPLICATION OF A FUGACITY MODEL FOR ASSESSING CHEMICAL FATE IN ECODISTRICTS OF SOUTHERN ONTARIO

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MANAGEMENT PERSPECTIVE

As part of a joint study between SOER and NWRI on environmental information analysis and integration for SOE Reporting, a regional toxic chemical distribution and fate model was used within the RAISON system. It was applied to ecodistricts of southern Ontario in order to estimate the relative distributions of selected chemicals between four bulk compartments as well as actual concentrations within the various ecodistricts. This type of modelling is very useful for directing monitoring and research activities towards the ecodistricts and compartments in which concentrations of contaminants are expected to be of significant concern. This can be used for chemicals which were released in the past, which are currently being released, and new chemicals can be evaluated for their potential to cause environmental problems before they are approved for use.

ABSTRACT

A fugacity model that was developed to assess the chemical fate of organic chemicals in regions of Canada has been incorporated into the RAISON (Regional Analysis by Intelligent Systems ON a microcomputer) expert system and has been modified and applied to ecodistricts of southern Ontario, Canada. The model is used to estimate a selected chemical's distribution between four bulk compartments (air, water, soil, and sediment) and 4 subcompartments (groundwater, coastal water, plants and animals). Polychlorinated biphenyls (PCB's), mirex, dieldrin, and hexachlorobenzene (HCB) were the chemicals considered in the application of the modelling system. Steady state concentrations predicted by the model were compared with available measured data for the ecodistricts. Considering the large degree of lumping required for ecodistrict parameters, the model predicts concentrations within the compartments that are reasonable estimates, especially considering the fact that concentrations between the compartments vary over as much as 10 orders of magnitude.

INTRODUCTION

Fugacity models have been used in a wide range of applications (Mackay 1991) and a special version was developed and tested for Health and Welfare (Mackay et al. 1991) to assess the fate of chemicals in Canada on a regional basis. As part of a pilot study between the National Water Research Institute (NWRI) of Environment Canada and State of Environment Reporting, the regional fugacity model (Mackay et al. 1991) has been incorporated into the RAISON expert system (Lam and Swayne 1992) and has been modified and applied to ecodistricts of southern Ontario. The model is used to estimate a selected chemical's distribution between four bulk compartments (air, water, soil, and sediment) and 4 subcompartments (groundwater, coastal water, terrestrial plants, and terrestrial animals).

Polychlorinated biphenyls (PCB), mirex, dieldrin, and hexachlorobenzene (HCB) were the chemicals considered in the application of the modelling system. The model is designed to consider the basic physical and chemical properties of the ecosystem and the basic chemical properties of the contaminant. The relative distributions and concentrations are controlled by the chemical's fundamental partitioning characteristics, its persistence in the environment and by the actual quantities or emissions discharged into the different compartments or media. This type of modelling approach is useful for directing monitoring and research activities towards the ecodistricts and compartments in which concentrations of contaminants are expected to be of significant concern. This would be the case for chemicals used in the past as well as for those that are currently being used. In addition, new chemicals can be evaluated for their potential to cause environmental problems before they are approved for use. The steady state version of the model considered in this study is designed to determine equilibrium concentrations that would exist after extended use of a chemical over a large region. A different modelling approach would be required to examine concentrations that would exist for specific sites which are affected by abnormally high discharges resulting from accidental spills or unregulated activities.

CONCEPTUAL MODEL

The model is based upon the fugacity concept (Mackay and Paterson 1981). Fugacity is a thermodynamic quantity related to chemical potential or activity that characterizes the escaping tendency of a chemical from a phase. At equilibrium, fugacities (units of pressure) are equal. It can be related to concentration in the same manner as temperature (°C) can be related to heat concentrations (cal/m³) using a proportionality constant, obtaining heat capacity (cal/[m³.°C]). The relationship for concentration and fugacity is:

C = Zf

where: f = fugacity (atm)

Z = fugacity capacity (mole/m³ atm)

C = concentration (mole/m³)

From this expression it can be seen that chemicals will tend to accumulate in phases where Z is high. In order to calculate how a chemical will partition, Z values must be determined for each bulk phase. When equilibrium exists between two phases, the fugacities are equal and the partitioning can be described by the ratio of their Z values. The derivation of the fugacity capacities for the different phases can be found in the original report (Mackay et al. 1991). The model considers eight compartments: 1) air, 2) water, 3) soil, 4) sediment, 5) ground water, 6)coastal water, 7) terrestrial plants, and 8) terrestrial animals. A schematic of the model is presented in Figure 1. It shows the four primary compartments of air, water, soil, and sediment. Partitioning equilibrium is assumed to exist within, but not between the compartments. There are discharges to air, water and soil and background inflows to air and water. As the media are internally homogeneous, it does not matter whether a chemical enters a media by discharge or flow, so there are only three total chemical input rates. There are seven overall intermedia transfer processes that represent a total of 20 individual transfer processes as it is

inconsequential whether the chemical is added or removed by advection, burial or transformation as long as it is permanently removed from the media.

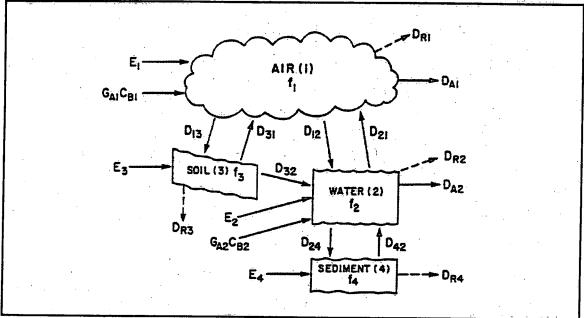


Figure 1 Fugacity model schematic

All rates of chemical transport and transformation are expressed as products of fugacities and D values which are transfer coefficients with units of (mole/h.Pa). In this model, there are seven intermedia D values which are expressed as functions of the horizontal areas of the media, media Z values, and a series of 15 constants which are combinations of transport terms such as mass transfer coefficients, diffusivities, and flowrates, all of which have units of velocity (m/h). The derivation of these D values are very lengthy and can be found in the original model development report (Mackay et al 1991).

Degradation reactions in the model are represented by first order expressions. These reactions are chemical specific and include processes such as photolysis, hydrolysis, oxidation and biodegradation. These rates are in the form of half-lives and when a chemical is subject to several reactions, a total half-life is used which represents the net effect of all of the processes.

The "ecopolitical" zones used in the original study have been replaced by ecodistricts. The current model application has been focused on the Great Lakes basin,

which was the Ontario mixed wood plain region of the original study (Mackay et al. 1991). The ecodistricts are presented in Figure 2.

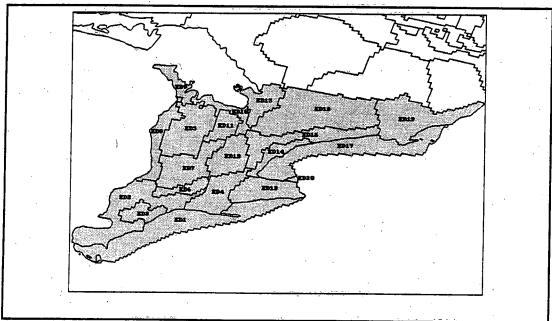


Figure 2 Ecodistricts of Ontario Great Lakes Basin, mixed wood plain region

The input data files required to run the model are presented in Tables 1 and 2.

Table 1. Ecodistrict Input File

- 1) Air Temperature mean summer, winter, and year values (°C)
- 2) Volume fraction of air particles
- 3) Volume fraction of water particles
- 4) Volume fraction of fish
- 5) Volume fraction of pure soil, soil air, soil water, and pore water
- 6) Volume fraction of sediment solid and pores
- 7) Total land area and water area
- 8) Precipitation (m/y)
- 9) Runoff (fraction of precipitation)
- 10) Air mass height (m)
- 11) Mean water, soil, sediment, groundwater, coastal water, plant and animal depths (m)
- 12) Bulk densities of the solid phases (kg/L)

- 13) Fraction organic content of solid phases
- 14) Advective flow rate (m³/h)

Table 2. Chemical Input File

- 1) Name of chemical
- 2) Molecular weight (g/Mol)
- 3) Vapour pressure (Pa)
- 4) Solubility (aq)
- 5) Log (octanol-water coefficient)
- 6) Reference temperature (°C)
- 7) Melting Point (°C)
- 8) Half-lives in air, water, soil and sediment (h⁻¹)

Additional input data which must be entered interactively during the execution of the model include the emissions (kg/y) into the bulk compartments of the 20 ecodistricts. These include both point source and non-point sources. Typically values are only entered for the air, water, and soil compartments. Emissions data for the chemicals were obtained from a wide range of sources (Swackhammer and Armstrong 1986, Strachan and Huneault 1979, Van Hove Holdrinet et al. 1978, Warry and Chan 1981, International Reference Group 1978, Halfon and Schito 1993, U.S. EPA and Environment Canada 1987, Frank et al. 1976) and represent the mean values for the period 1950-1990.

The integration methods used to obtain the data required for the physical characteristics of the ecodistricts are similar to those used in the original fugacity model report for ecoregions (Mackay et al. 1991). Actual values for the ecodistrict physical characteristics were obtained from several sources. Air temperatures and precipitation data were obtained from the Great Lakes Atlas and Resource Book (U.S. EPA 1987).

Total land areas and water areas were obtained using GIS area analysis methods of digital maps. Soil properties for each ecoregion are based on spatial averages of information from Agriculture Canada reports for the different counties and these data were converted to the ecodistrict boundaries using algorithms built into the RAISON system. The input data required for the chemicals were obtained from several sources (Mackay et al. 1991, Mackay et al. 1992).

SENSITIVITY ANALYSES

Sensitivity analyses were carried out on a "generic" version of the model for the 4 chemicals using a program supplied by the model developer (Mackay et al. 1991). The sensitivity, S_{ji} is the ratio of the relative change in concentration C_i in each of the four bulk media (air, water, soil, and sediment) to the relative change in each of the 10 parameters (X_i) listed in Table 3. The program was run for each chemical assuming that emissions into air, water, and soil were equal and that there was no additional inflow concentration in the air or water. Sensitivity was calculated independently for each parameter for a 5% increase. As the model is linear, results would be the same if a different increase or decrease was applied to all parameters. In Table 3 the results are shown in ascending order of absolute value for the top 20 of the 40 independent values of S_{ji}. It can be seen that each of the chemicals have different sensitivity results. For example, for PCB, the greatest relative changes in concentration occur in soil (C₃) when its emission in that medium (X₁₀)) or half-life (X₆) is varied. For dieldrin, the greatest relative change in concentration occurs in air (C1) when its emission into air is varied. The greatest relative change for mirex concentrations in sediment, soil, water, and air occur when emissions into water (X₉) are varied. For hexachlorobenzene, the greatest relative change in concentration occurs in sediment (C4) when its octanol-water partition coefficient (X₃) in that medium is varied.

	PCB		Table 3	Sensitivity Dieldrin	3 Sensitivity Analysis of Regional Fugacity Model Parameters Dieldrin	gional Fugac	ity Model Pa	arameters	n n	levach Orchangene	9
Sensitivity Parameter	×	S,	Sensitivity Parameter	×	S _{j.}	Sensitivity Parameter	×	Š	Sensitivity Parameter	X	Si
S33	k,soil	1.996E-03	S	vp-soil	1.406E-02	Š	vn-water	3 \$05E-03	v	rio o	1 4185 00
S ₄₃	hl,-soil	2.472E-03	S	hl,-sed	1.491E-02	S	vp-sed	3.507E-03	<u> </u>	VD-air	5 200E-02
S ₁₀₁	e3-air	2.509E-03	Seg	hlawater	1.491E-02	S.	sol-sed	3.668E-03	S.	sol-air	5.328F-02
S ₆₁	hl ₃ -air	2.509E-03	Sig	e ₃ -sed	1.492E-02	, S	sol-water	3.669E-03		sol-soil	7.815E-02
S ₈₃	e ₁ -soil	2.511E-03	S ₁₀₂	e ₃ -water	1.492E-02	S_{21}	vp-air	2.103E-02	လို	vp-soil	8.342E-02
S _o	e ₂ -air	2.568E-03	S ₈₃	e ₁ -soil	1.584E-02	Sıı	sol-air	2.202E-02	S	ksoil	9.047E-02
Sel	hl ₂ -air	2.572E-03	S44	hl ₁ -sed	1.718E-02	S_{32}	k _w -water	8.930E-02	Š	kair	9.316E-02
S,	hl,-air	2.804E-03	S _{42.}	hl ₁ -water	1.718E-02	S_{33}	k _{ow} -soil	1.644E-01	Š	kwater	1.349E-01
	k _{ow} -air	3.275E-03	S_{21}	vp-air	2.392E-02	Sz	k _{ow} -sed	2.209E-01	S	e ₁ -air	1.364E-01
S ₁₁	sol-air	3.322E-03	S _z	e1-sed	2.672E-02	Ss	hl ₂ -sed	4.138E-01	S ₂₂	vp-water	1.713E-01
N C	k _{ow} -sed	1.471E-01	S ₈₂	e ₁ -water	2.672E-02	S _{S1}	hl ₂ -air	4.138E-01	S	vp-sed	1.713E-01
ν 1	hl ₁ -air	9.474E-01	S ₅₂	hl ₂ -water	5.361E-02	S _{S2}	hl ₂ -water	4.138E-01	S	sol-sed	1.787E-01
S ₇₄	hl,-sed	9.868E-01	SS	hl ₂ -sed	5.361E-02	S_{53}	hl ₂ -soil	4.138E-01	S	sol-water	1.787E-01
S	hl ₂ -sed	9.905E-01	S ₄₁	hl ₁ -air	6.430E-01	S ₁₃	sol-soil	6.105E-01	S.	hl,-air	6.227E-01
S ₅₂	hl ₂ -water	9.905E-01	S ₇₄	hl₄-sed	7.892E-01	S_{23}	vp-soil	6.490E-01	လို	e,-air	8.494E-01
S	e ₁ -air	9.965E-01	Sr	e ₂ -sed	9.584E-01	S ₃₁	k, -air	7.660E-01	S	hi ₁ -soil	8.787E-01
. S	e ₂ -water	9.996E-01	S ₉₂	e ₂ -water	9.584E-01	S	e ₂ -sed	1.000E+00	S	e ₂ -sed	9.957E-01
a S	cz-sed	9.996E-01	Seg	hl ₃ -soil	9.837E-01	S ₉₃	e ₂ -soil	1.000E+00	S	e ₂ -water	9.958E-01
, S	hl ₃ -soil	1.001E+00	S ₁₀₃	e ₃ -soil	9.842E-01	S ₉₂	e ₂ -water	1.000E+00	S ₁₀₃	e ₃ -soil	9.962E-01
S ₁₀₃	e ₃ -soil	1.001E+00	S ₈₁	e ₁ -air	1.000E+00	S91	e ₂ -air	1.000E+00	Szd	k _{ow} -sed	1.223E+00
Parameters (X _j) 1. Vapour Pressure 2. Solubility 3. Octanol/water partiti 4. Half-life in air 5. Half-life in water 6. Half-life in soil 7. Half-life in sediment 8. Emissions into air 9. Emissions into water 10. Emissions into water	ameters (X _j) Vapour Pressure Solubility Octanol/water partition coefficient Half-life in air Half-life in soil Half-life in soil Emissions into air Emissions into air Emissions into soil	efficient		Media Concentrations (C _i) 1, Air 2. Water 3. Soil 4. Sediment	ntrations (C,)			Symbols hi, = half-life for reason = water solubility vp = vapour pressure e = emission rate		edia i	

RESULTS

The model has been applied to the 20 ecodistricts for 4 different chemicals: 1) Polychlorinated biphenyl (PCB), 2) mirex, 3) dieldrin, and 4) hexachlorobenzene (HCB). PCB's have been manufactured since the late 1920's and have been in use in the Great Lakes Basin for more than 40 years. Mirex (dodecachlor-octa hydro-1,3,4-metheno-2H-cyclo-buta (c.d) pentalene) was produced by Hooker Chemicals on the east bank of the Niagara River between 1959 and 1976. It was used as a flame retardant and as a military pyrotechnic under the trade name Dechlorane and as the active ingredient to kill fire ants in the southern USA. It was known to enter the Great Lakes Basin via the Niagara and the Oswego rivers.

The pesticide aldrin, which is the parent compound of dieldrin, was used in the Great Lakes Basin from the 1950's until 1969 in Ontario and 1974 in the United States. HCB is used in the plastics industry and in the manufacture of dyes. It is very stable in the environment and has been shown to be carcinogenic in laboratory tests.

The distribution of a chemical within an ecodistrict is available for each of the 8 compartments. Examples of the distributions are shown in the form of pie charts for a number of ecodistricts. In all of the examples, the results are for mean summer conditions and the emissions used represent mean values for the period that the chemical has been in use. The percentage distributions represent the fraction of the total number of moles of contaminant in each ecodistrict. In Figure 3 the distributions of PCB's are shown for ecodistricts 1, 2, and 13. Ecodistrict 1 represents Lake Erie and it can be seen that virtually all of the mass of PCB is associated with the lake sediments. In ecodistrict 2, the PCB's are predicted to be tied up in the soil, and plant and animal biomass. In ecodistrict 13, 65% is distributed within the plants and animals and the remainder is equally distributed between the soil and bulk sediments.

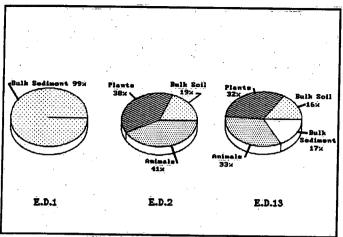


Figure 3 PCB compartmental distribution (% of total moles for the ecodistrict)

In Figure 4 the distributions of dieldrin are shown for ecodistricts 1, 2, and 13. As compared to PCB in ecodistrict 1, it can be seen that not all of the chemical is expected to be tied up with the sediment. The majority is still associated with the sediments but 11% is now expected to be found within the plants and animals. In ecodistrict 2, the same distribution as seen for PCB is predicted.

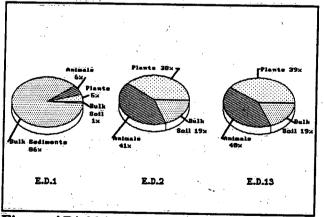


Figure 4 Dieldrin compartmental distribution (% total moles in the ecodistrict)

The distributions predicted for mirex in ecodistricts 1, 2, and 13 are shown in Figure 5. It can be seen that mirex behaves similarly to PCB's in ecodistrict 1, being associated almost exclusively to the lake sediments. In ecodistrict 2, the distribution is again very similar to that of PCB's, except that 3% is now predicted to reside with the lake sediments. In contrast, in ecodistrict 13, the percentage of mirex predicted to be tied up with lake sediments is much lower than is predicted for PCB's. This points out that it is not just the chemical behaviour of the contaminant that controls its distribution but also the physical and chemical nature of the ecodistrict components.

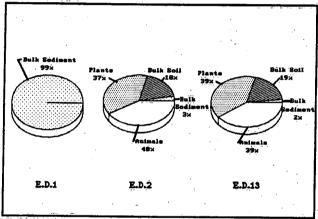


Figure 5 Mirex distribution (% total moles)

The distributions of HCB within ecodistricts 1, 2 and 13 are shown in Figure 6. As for PCB and mirex, almost all of the HCB is expected to be associated with the lake sediments. In contrast to PCB and mirex distributions in ecodistricts 2 and 13, 85% of the HCB is predicted to be associated with lake sediments and only 5% is to be expected in plants and animals as compared to 40% for PCB and mirex. The key reason for the lack of HCB in plants and animals is that HCB has a much lower affinity for lipids which are the compounds in plants and animals that hydrophobic compounds such as PCB and mirex preferentially partition into. Comprehensive measured data do not exist for the different ecodistricts for each of the compartments to verify the model results with respect to total distribution of mass. However, measured concentrations are available from several independent studies which can be used to compare with model predictions.

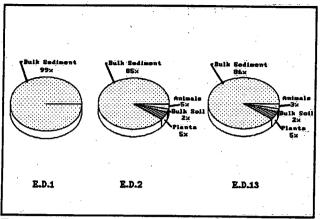


Figure 6 HCB distribution (% total moles)

In order to actually compare model predicted concentrations with observed data, results are shown as bar graphs on the Ontario Great Lakes ecodistrict map. In all of the examples shown it will be seen that measured data are not available for many of the ecodistricts. The measured data are also usually only available for a single point for a specific compartment component, which is not necessarily representative of the compartment as a whole, as it is considered in the model.

In Figure 7 the measured vs predicted concentrations of PCB in bulk air are presented. The scale is logarithmic as the measured and predicted concentrations range over several orders of magnitude. The units of concentration are ppb. It should be pointed out here that this type of range in measured values is typical for these types of contaminants. It can be seen that the measured values are consistently greater than the predicted values. In the model, the bulk air concentrations are calculated for a column of air 2000 metres in height. Measured values are typically for heights of only a few metres above the ground and consequently would be expected to be higher than observations at greater heights as the sources are at or near ground level.

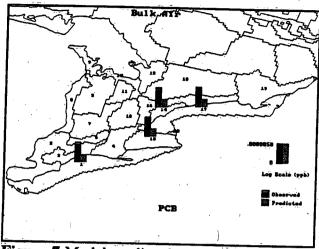


Figure 7 Model predicted vs measured bulk air concentrations (ppb)

The measured vs predicted concentrations of PCB in the bulk water compartments of various ecodistricts are shown in Figure 8. It can be seen immediately that the concentrations in the lakewater are approximately 3 orders of magnitude lower than those in the bulk air. Observed values are less than the predicted values for E.D.1 (Lake Erie) and E.D. 17 (Lake Ontario). In the other terrestrial ecodistricts, the observed values are greater than the predicted values. These differences are due mainly to the fact that the measured data are for a specific period in time (1976) and the emissions are the mean of emissions for the period 1950-1990. A time variable version of the model would need to be used to obtain more precise predictions.

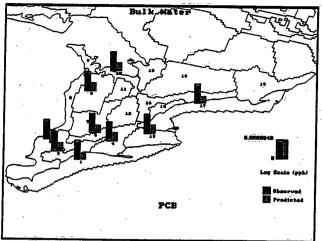


Figure 8 Model predicted vs measured bulk water concentrations (ppb)

In Figure 9 the measured and predicted concentrations of PCB in bulk soil are presented. Soil concentrations range from 1 to 10 ppb, which are 3-4 orders of magnitude greater than those in bulk water. Again, the differences between the observed and measured values fall within the uncertainties in the emissions used as input to the model.

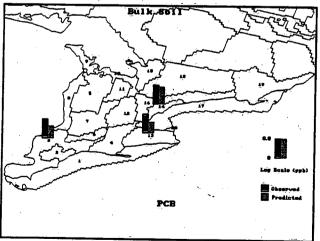


Figure 9 Model predicted vs measured bulk soil concentrations (ppb)

Bulk sediment concentrations are shown in Figure 10. The measured and predicted concentrations for the two ecodistricts for which measured data were available (E.D.1 and E.D.17) compare quite well.

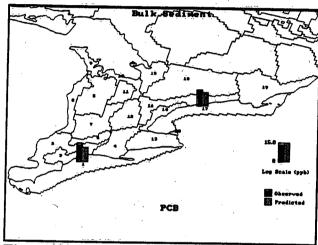


Figure 10 Model predicted vs measured bulk sediment concentrations (ppb)

In Figure 11, the concentrations in terrestrial animals are presented. The measured data represent the mean concentrations of PCB in herring gull eggs. The actual data show a continuing decrease in the concentrations since measurements started in 1974. This again points out that to more accurately predict concentrations in a compartment, a time-dependent version of the model would be required.

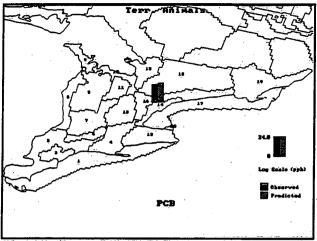


Figure 11 Model predicted vs measured terrestrial animal PCB concentrations (ppb)

Figures for the other three chemicals for the various compartments are included in the Appendix. Overall, the model has proven to be very useful in predicting the fate of chemicals within the limits that the model has been designed. Obviously due to the high degree of lumping and aggregation required to carry out regional analyses, it is not reasonable to expect the model to predict concentrations in specific compartment components accurately. This would require more detailed knowledge of the subcompartments and would best be modelled with a different version of the Fugacity model used in this exercise or another model designed for a more site specific type of application.

Future work would involve obtaining better emissions data as well as investigating other possible sources of observed data in order to more fully calibrate the steady-state model. Then it would be useful to apply the time-dependent version of the model to examine the effects of time-variable loadings and the associated trends in observed concentrations.

ACKNOWLEDGEMENTS

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APPENDIX

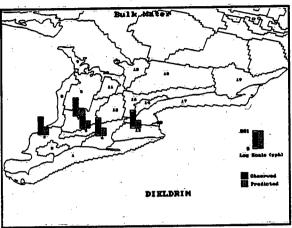


Figure 12 Model predicted vs measured bulk water dieldrin concentrations (ppb)

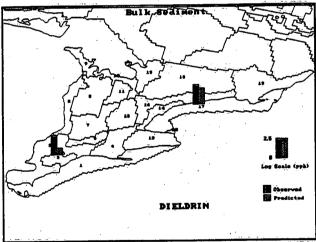


Figure 13 Model predicted vs measured bulk sediment dieldrin concentrations (ppb)

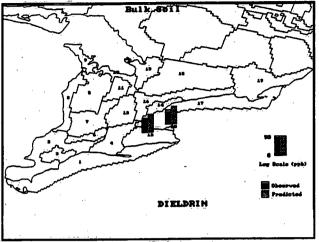


Figure 14 Model predicted vs measured bulk soil dieldrin concentrations (ppb)

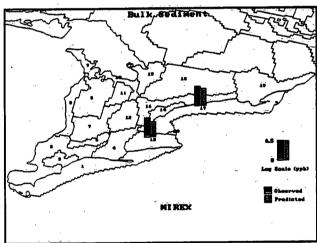


Figure 15 Model predicted vs measured sediment mirex concentrations (ppb)

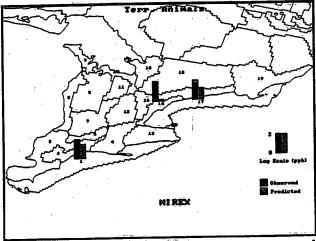


Figure 16 Model predicted vs measured terrestrial animal mirex concentrations (ppb)

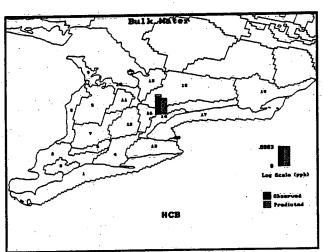


Figure 17 Model predicted vs measured bulk water HCB concentrations (ppb)

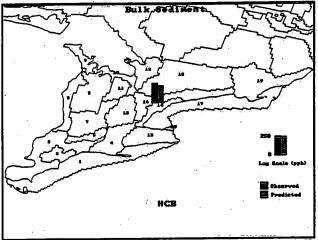


Figure 18 Model predicted vs measured bulk sediment HCB concentrations (ppb)

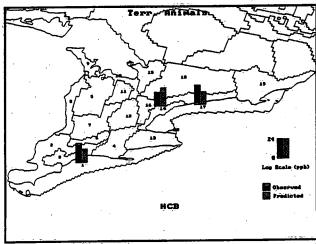
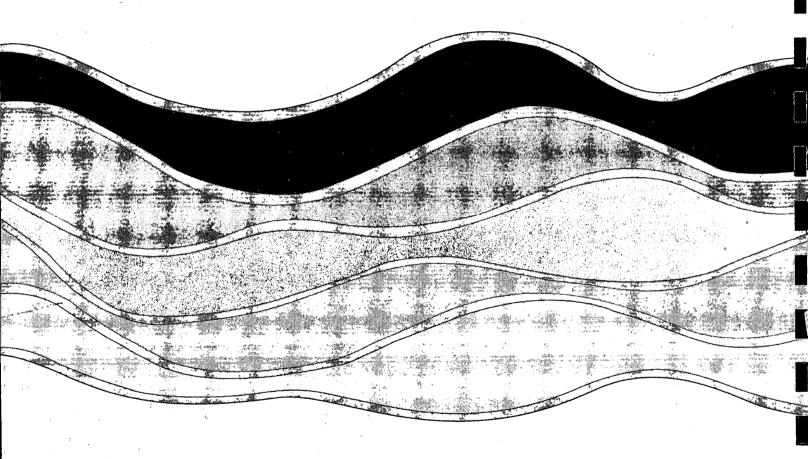


Figure 19 Model predicted vs measured terrestrial animal HCB concentrations (ppb)

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