OBT Formation in Night Experiments and Modeling Trials at CRL

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EMRAS II

- **IAEA** (International Atomic Energy Agency)'s Programme
- **E**nvironmental **M**odelling for **Ra**diation **S**afety II
  - Intercomparison and Harmonization Project
- 9 Working Groups in EMRAS II
  - Working Group 7 : "**Tritium**" Accident
    1) Two goals (Optimization and Uncertainty)
    2) Canada is one of the leading countries

Outline

- Background on environmental tritium in Canada
- Knowledge gaps in OBT formation
- HTO exposure experiments at night
- Plant physiology
- Conceptual and mathematical model
- Example of OBT prediction at night
- Summary

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Canada's Nuclear Power Reactors

- 17 CANDUs are currently operating
- 3 are being refurbished
- 2 are in guaranteed shutdown state

Gentilly, QC

Darlington, ON

Bruce, ON

Pickering, ON

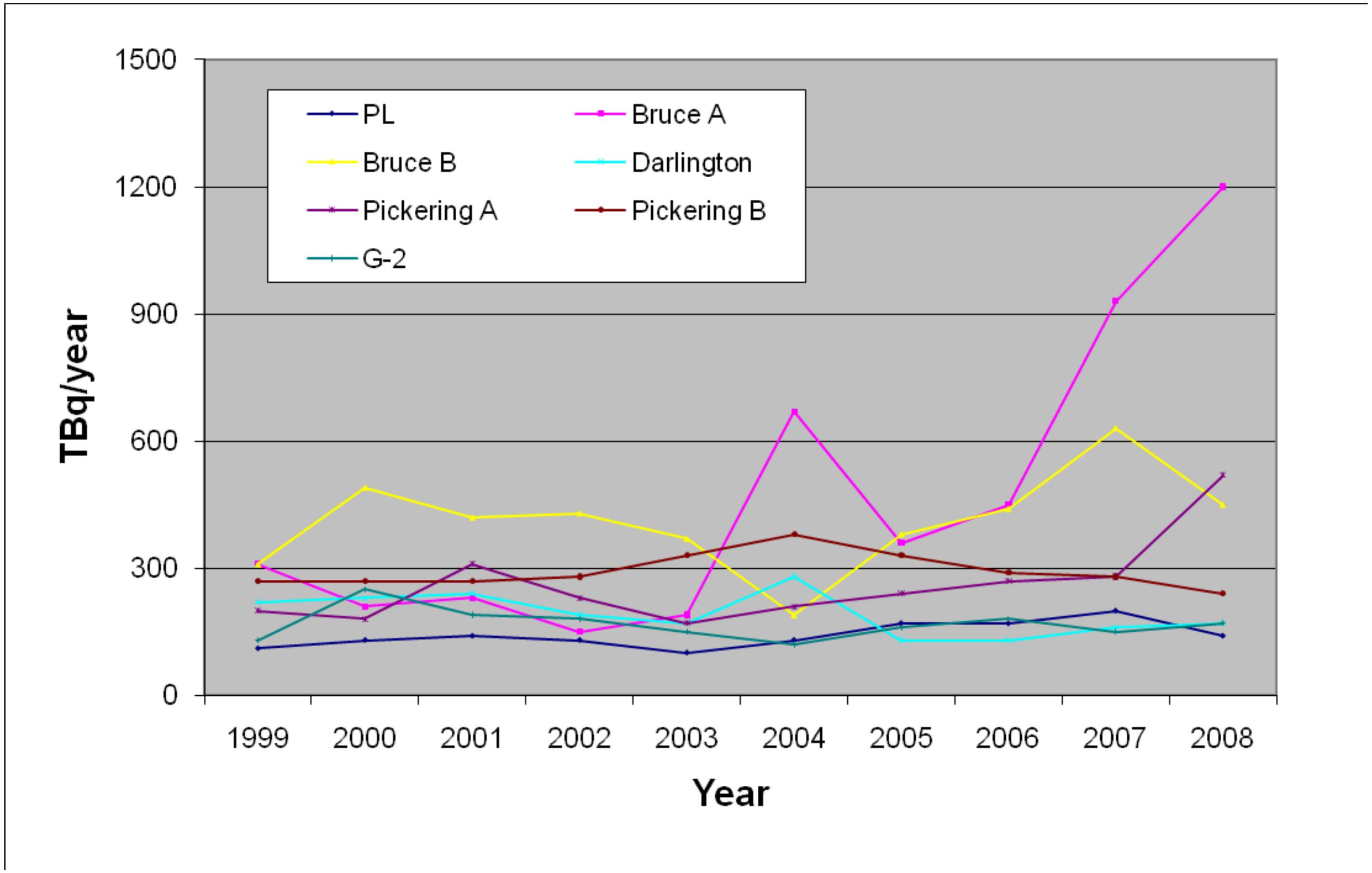
Pt. Lepreau, NB

Nuclear Facilities in Canada

Type	Location	Facilities	In Service
Nuclear Power Generating Station	Ontario	Darlington	1990
		Pickering	1971
		Bruce	1978
	Quebec	Gentilly-2	1983
	New Brunswick	Point Lepreau	1983
Tritium Processing Facilities	Ontario	SRB Tec.	N/A
		SS Inc.	N/A
Research Facilities	Ontario	CRL	1952
	Manitoba	WL	1963-1998

N/A = Not available

Tritium Oxide in Gaseous Effluent



Source from CNSC

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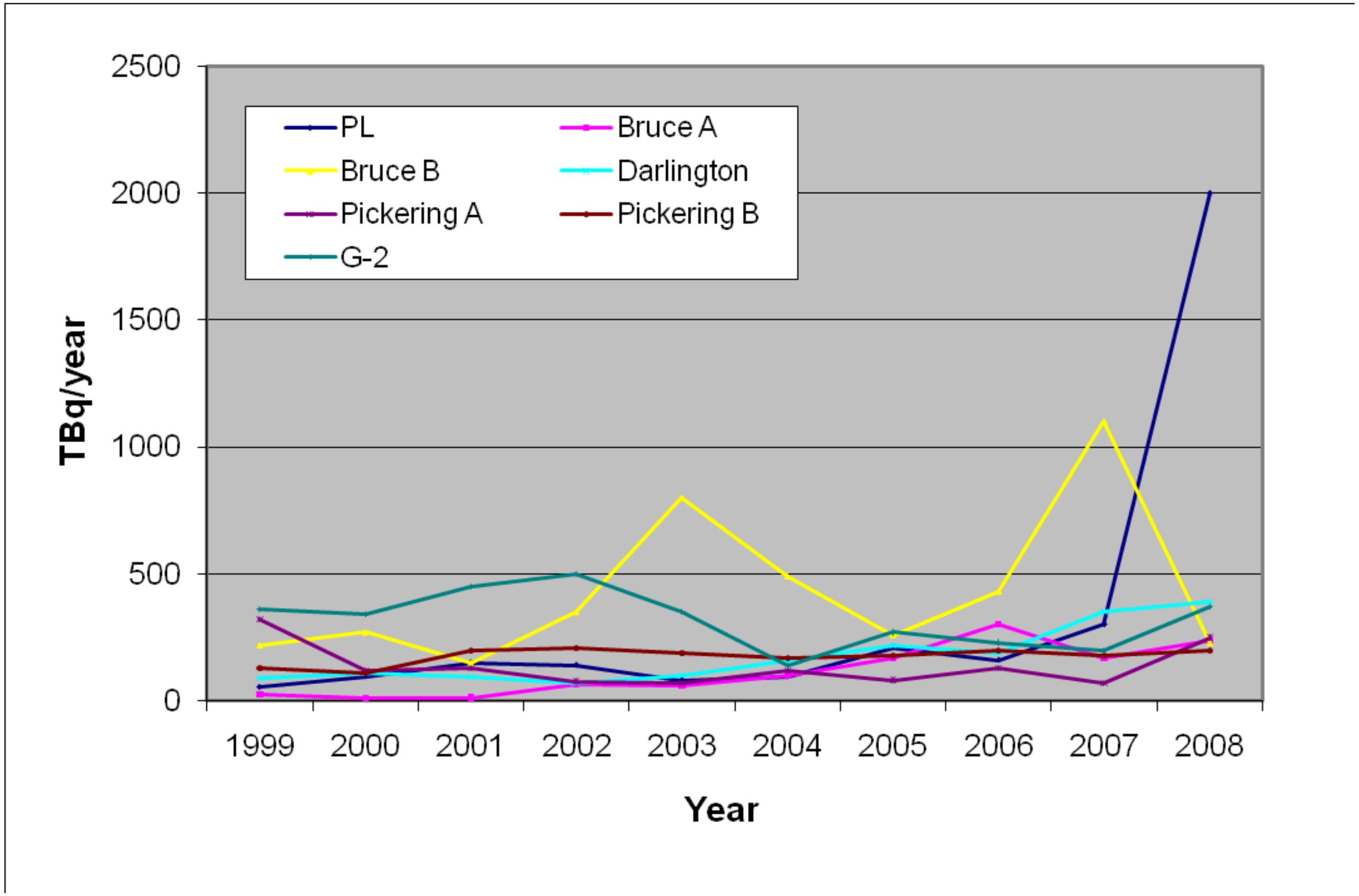
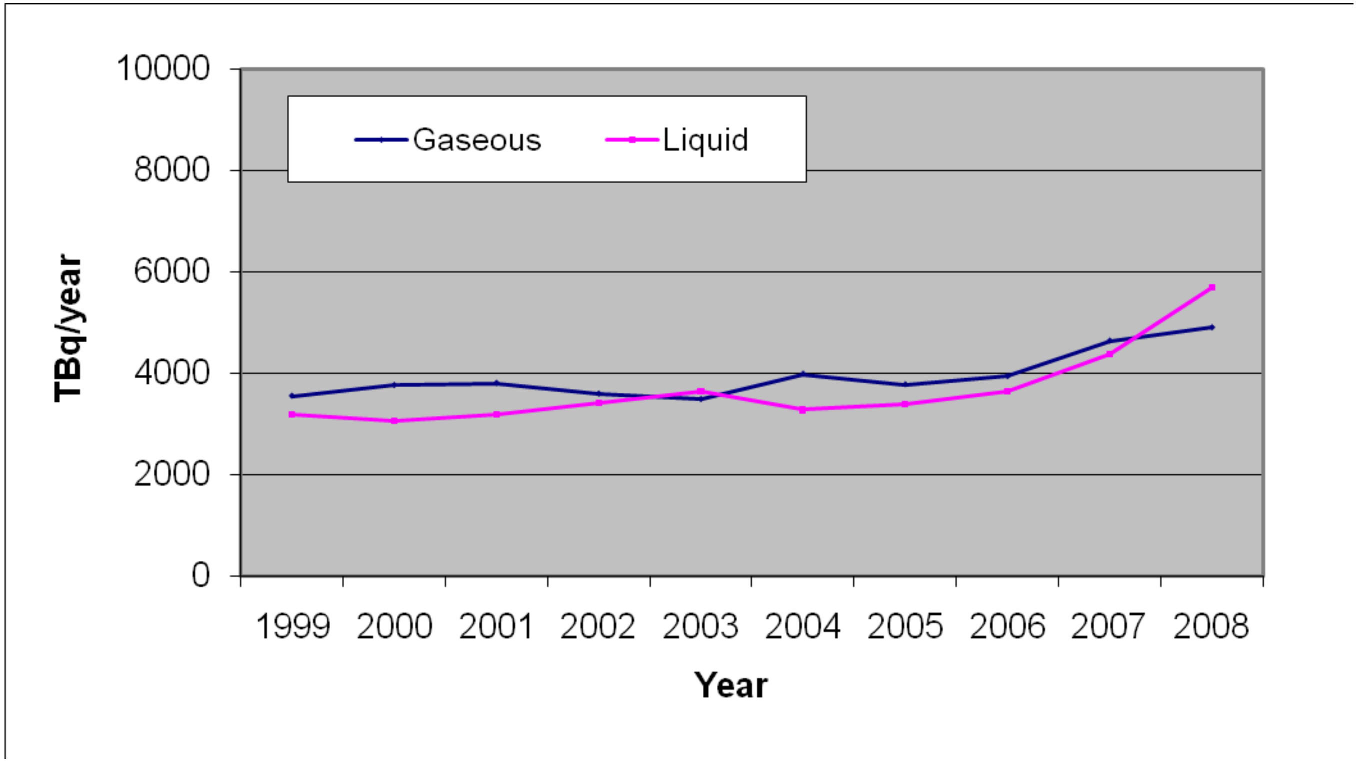
Tritium Oxide in Liquid Effluent



Total HTO in Released Effluents in Canada



What is the fate of tritium released into the environment?

Regulation (International Limits for Tritium in Drinking Water)

Countries/ Organization	Tritium Limit (Bq/L)	Application
Health Canada, Ontario and Quebec	7,000	Guideline Standard
U.S.A. EPA California EPA	740 15	Max. Contaminant L. Public Health Goal
European Union	100	Screening Value
Finland Australia	30,000 76,103	Standard Guideline
WHO	10,000	Guideline

The Ontario Drinking Water Quality Standard for tritium was revised to 20 Bq/L (2009)!

Environmental Issues

- Environmental release forms are HT and HTO
- Environmental measureable forms are HTO and OBT
- HTO measurement is relatively simple and straightforward
- OBT behaviour in the environment is relatively complicated and has a higher uncertainty than HTO behaviour
    - OBT measurement is useful for normal operations
    - OBT prediction is useful for accidental situations

Knowledge Gaps in OBT Formation

- Theory of OBT formation in plants and animals
- Fraction of exchangeable and non-exchangeable OBT
- OBT formation and translocation during the day
- OBT formation and translocation at night
- OBT behaviour in the terrestrial ecosystem
- OBT behaviour in the aquatic ecosystem
- Uncertainty of OBT measurement and OBT prediction

HTO Exposure Experiments at Night

- The first experiment was conducted in Germany (1996)
  - Open wheat field using an exposure chamber
- The second experiment was conducted in Korea (1998)
  - Rice pots using an exposure chamber
- The third experiment was conducted in Canada (2004)
  - Open field experiment with tomato pots at Perch Lake (2001)
  - Tomato pots using an exposure chamber (2004)

Experiments in Germany

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Experiments in Korea

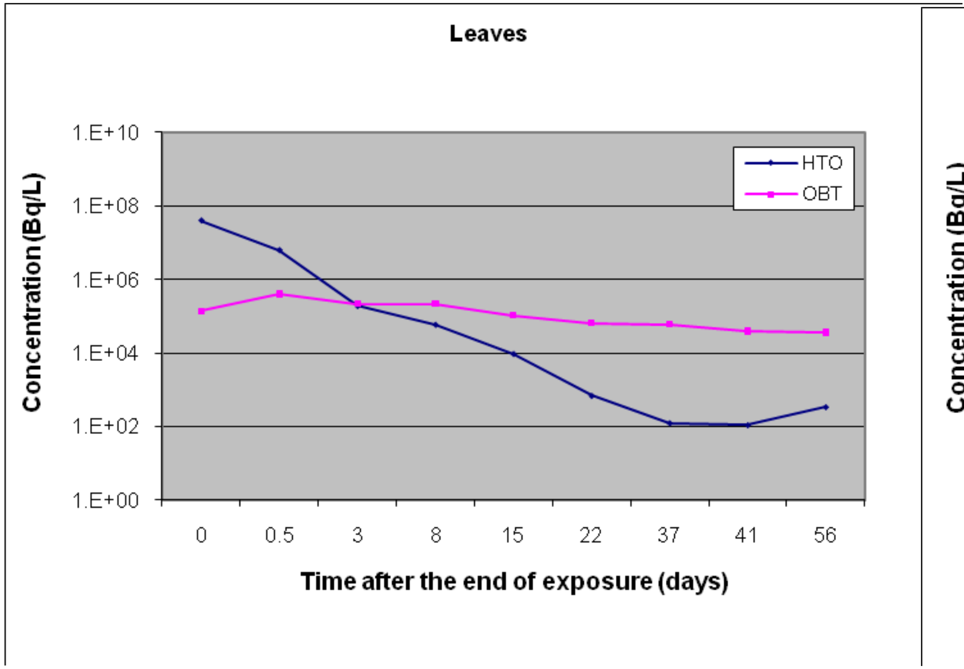
Two Different CRL Experiments

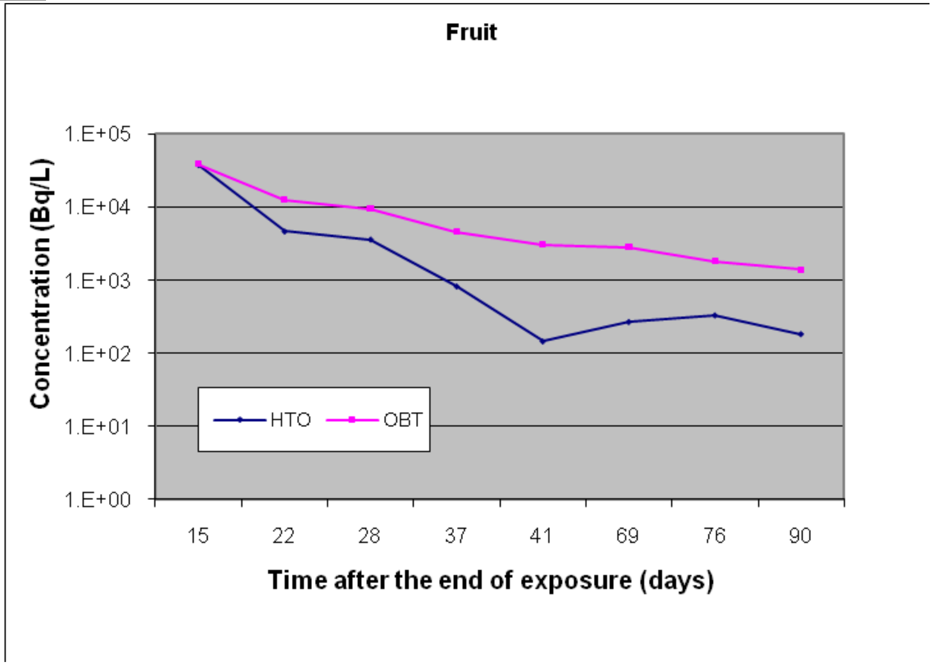
- Kotzer et al. (2001): Exposed potted tomato plants for short periods of time (7 or 8 hours) to elevated tritium concentrations at Perch Lake
- The experiment was not successful because the air concentrations were too low to induce detectable increases in the OBT concentrations in the plants
- To ensure the air concentrations were sufficiently high to obtain reliable results, the exposures were carried out in a chamber in which the air concentration should be brought to an arbitrarily high level

Experimental Conditions (2004)

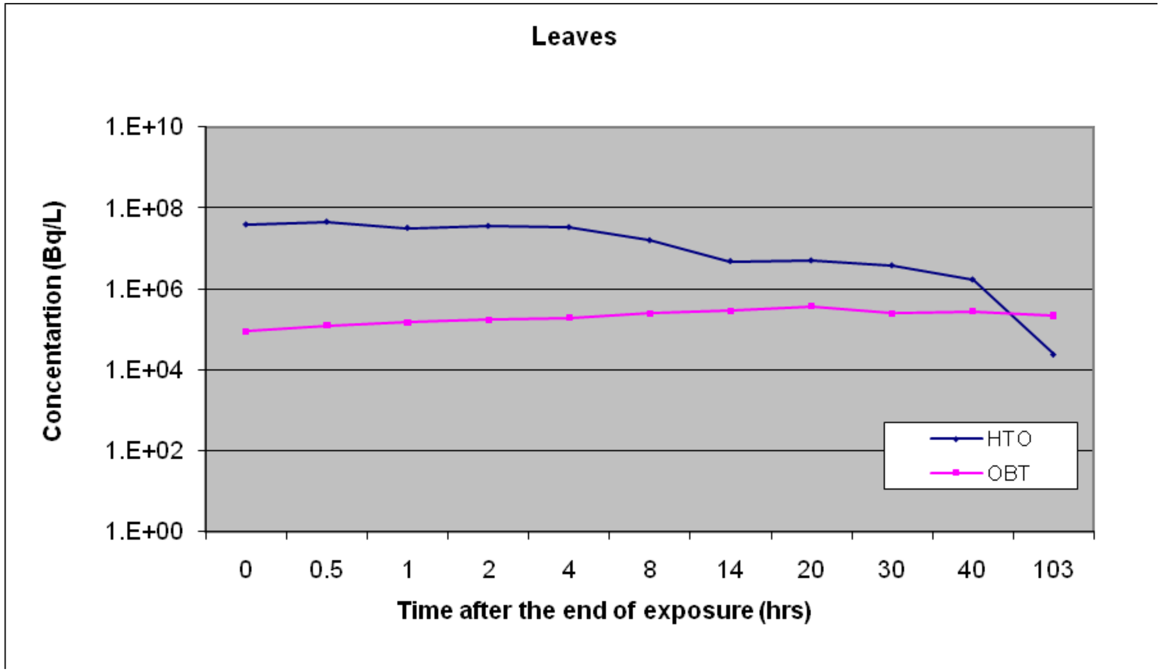
Date &Time	External conditions	Growth stage	Sampling protocol
July 6 (22:00)	Clear & 16ºC	Early (no fruit)	Long term (2 plants)
July 11 (22:00)	Cloudy & 22ºC	Early (no fruit)	Short term (2 plants)
July 21 (22:00)	Cloudy & 26ºC	Intermediate (green fruit)	Long term (2 plants)
July 22 (21:30)	Clear & 27ºC	Intermediate (green fruit)	Short term (2 plants)
Aug 23 (21:00)	Clear & 15ºC	Late (ripe fruit)	Long term (1 plant)
Aug 24 (21:00)	Clear &17ºC	Late (ripe fruit)	Short term (1 plant)

Time Variation of HTO and OBT (Exp 1)



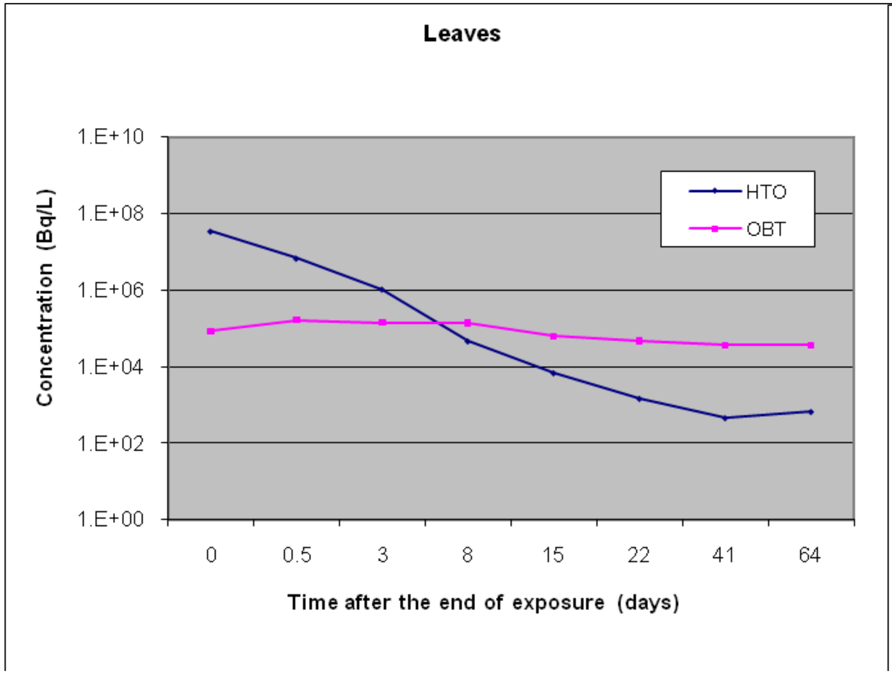


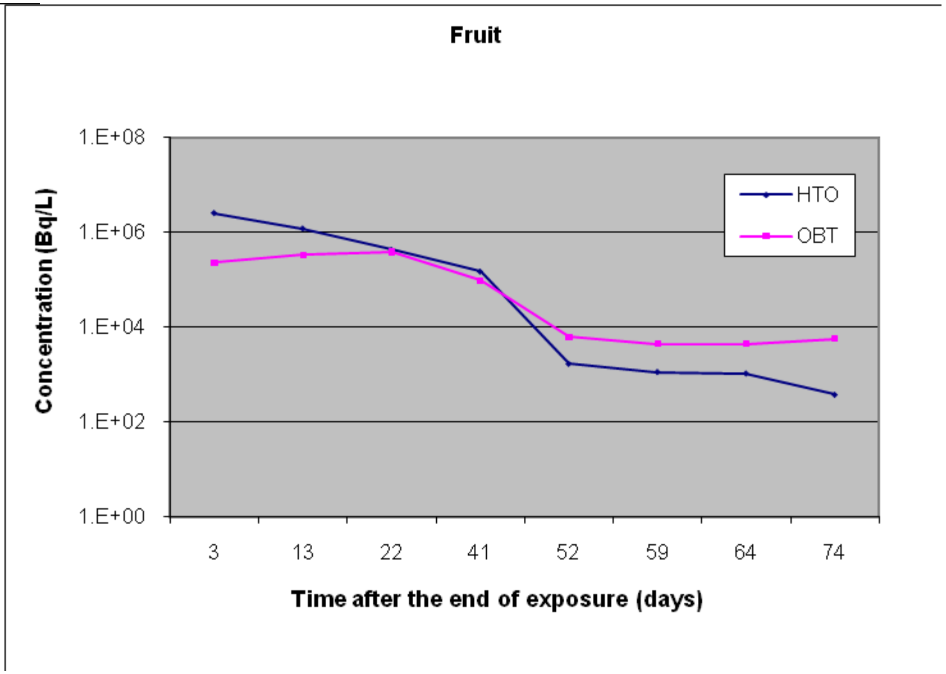
Time Variation of HTO and OBT (Exp 2)



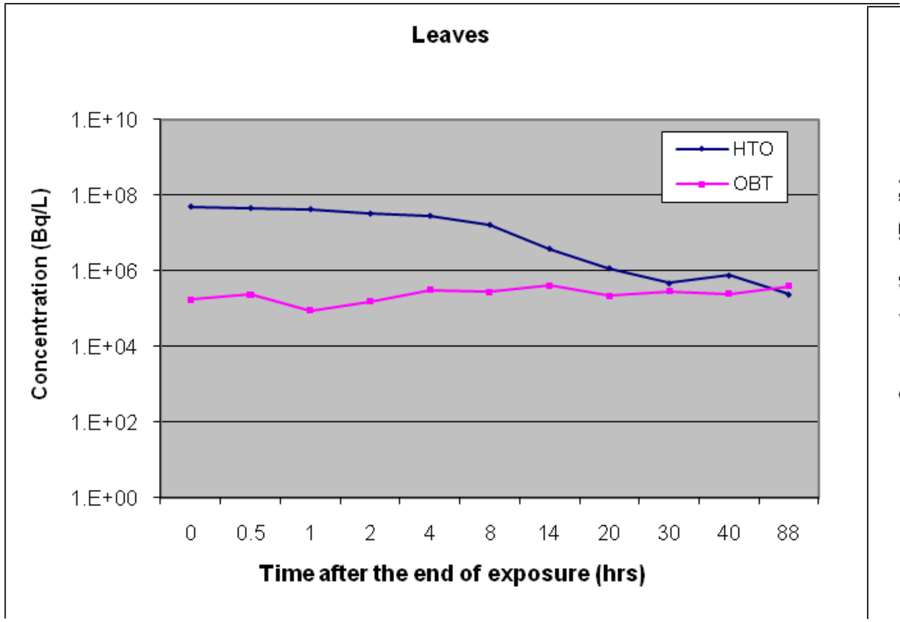
Good agreement with Exp 1

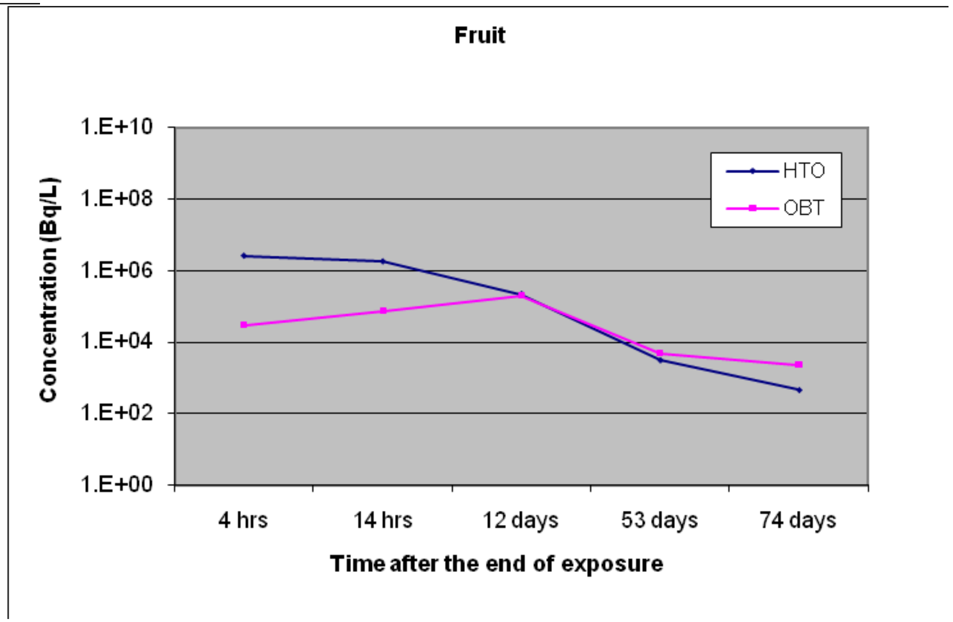
Time Variation of HTO and OBT (Exp 3)



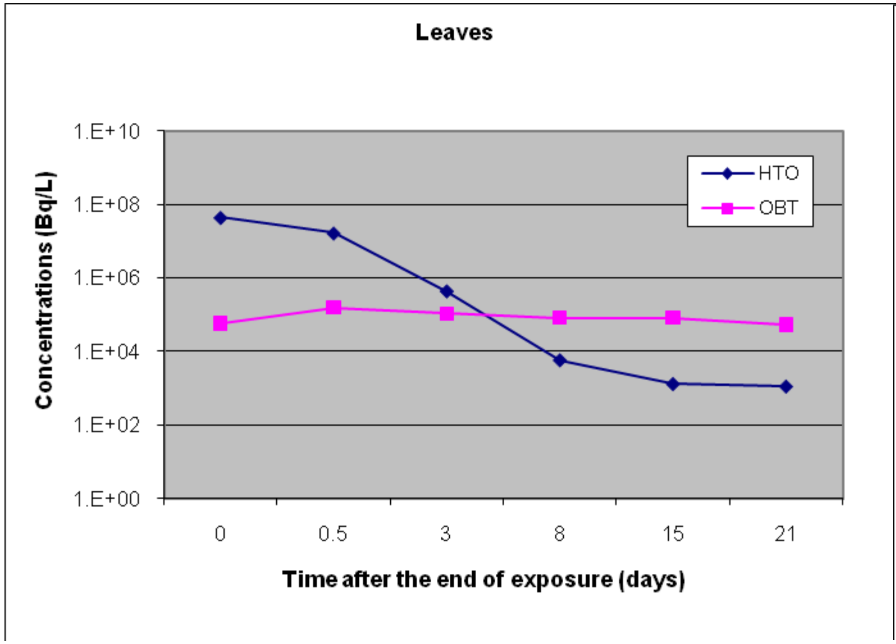


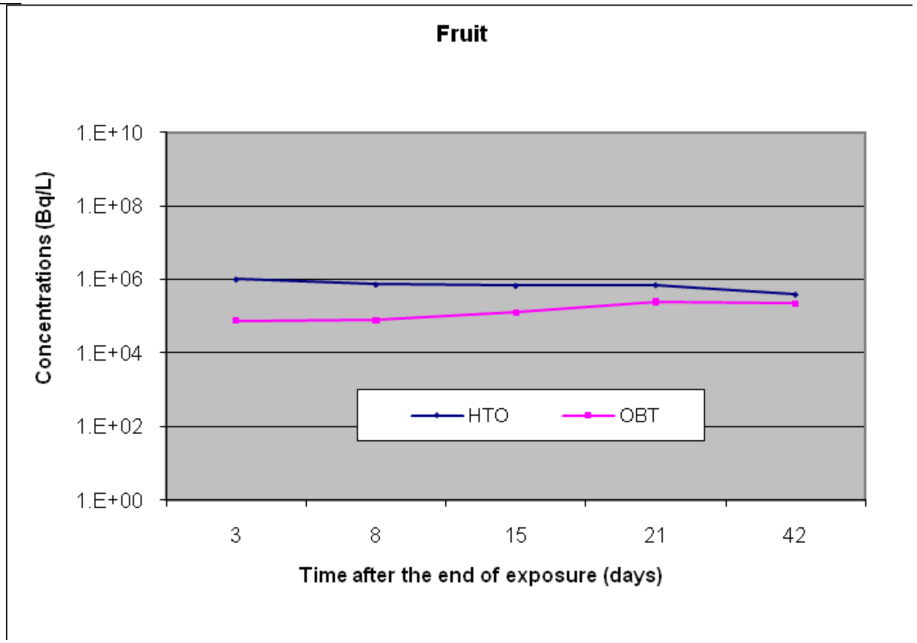
Time Variation of HTO and OBT (Exp 4)





Time Variation of HTO and OBT (Exp 5)





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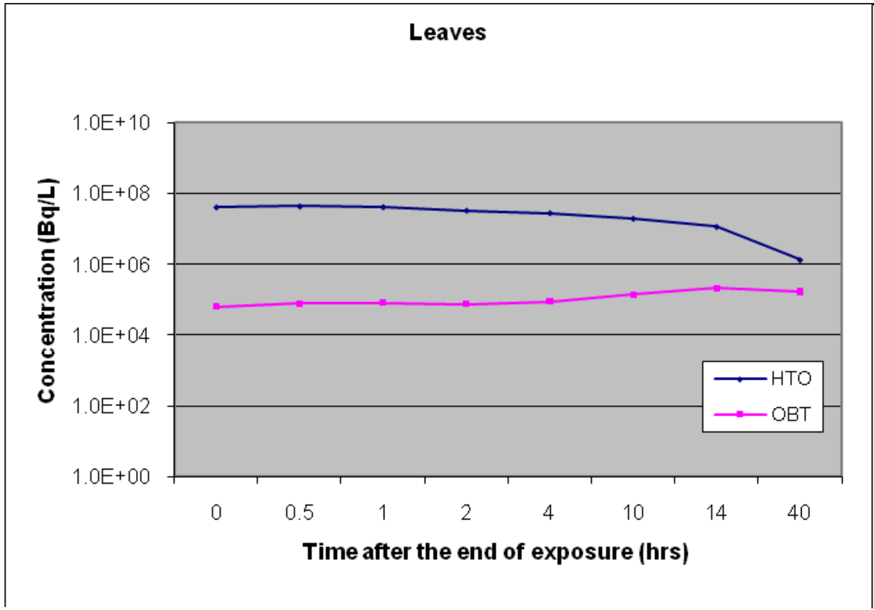
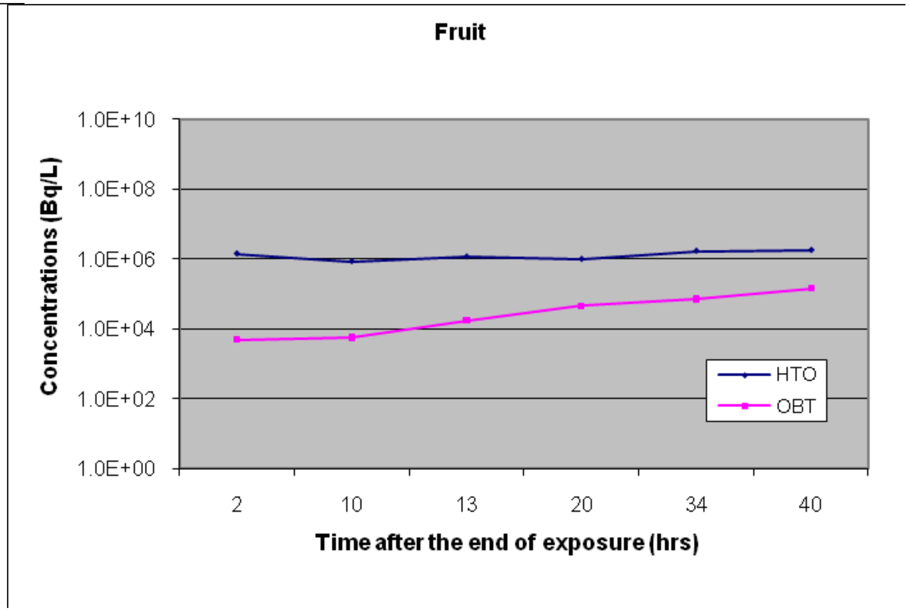
Time Variation of HTO and OBT (Exp 6)





Maximum HTO and OBT Concentrations in Leaves

Exp	Max. OBT (Bq/L)	Time (hrs)	HTO (Bq/L)	Type
1	4.06×10^5	12	3.96×10^7	Night
2	3.66×10^5	20	3.79×10^7	Night
3	1.66×10^5	12	3.48×10^7	Night
4	4.15×10^5	14	4.97×10^7	Night
5	1.58×10^5	12	4.43×10^7	Night
6	2.08×10^5	14	4.13×10^7	Night
7	5.06×10^5	6	5.27×10^7	Day
8	5.30×10^5	2	5.74×10^7	Day

Measured OBT Formation Rates in Fruit

Exp.	Interval (h)	R_f Rate (h^{-1})	Interval (h)	R_m Rate (h^{-1})
1	361	2.76×10^{-6}	-	-
2	-	-	-	-
3	73	8.97×10^{-5}	529	2.09×10^{-5}
4	5	1.21×10^{-4}	-	-
5	73	2.32×10^{-5}	505	1.10×10^{-5}
6	6	3.90×10^{-5}	-	-

R_f is the rate calculated from the start of exposure to the time of the first OBT measurement

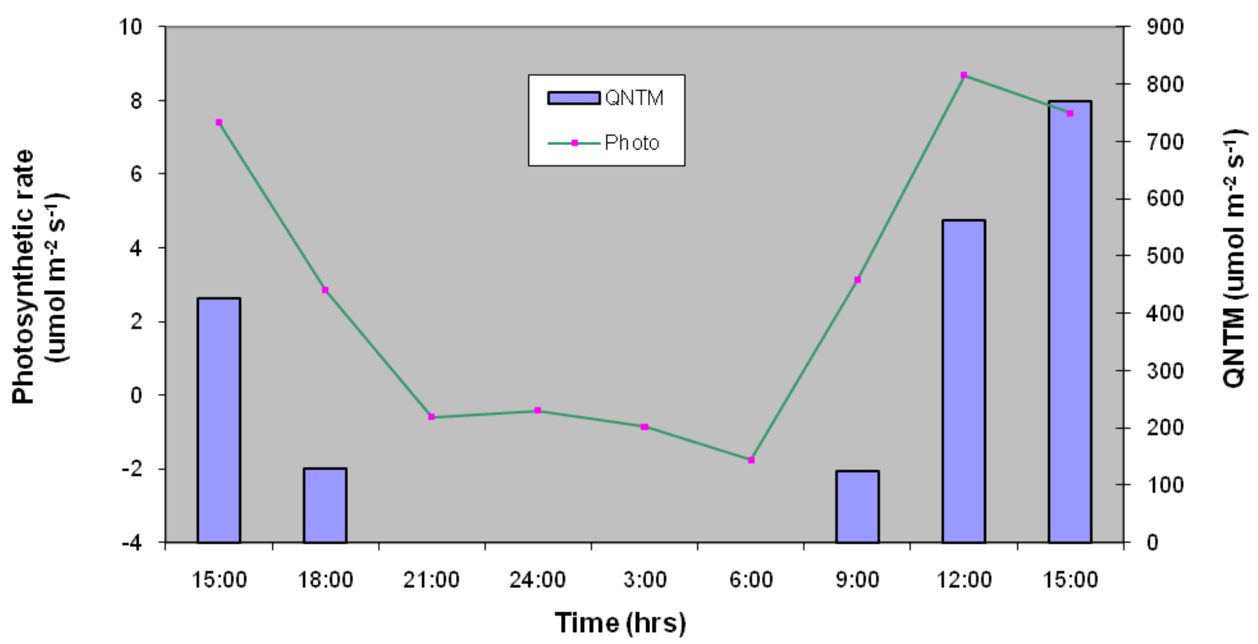
R_m is the rate calculated from the start of exposure to the time of the maximum OBT concentration

Plant Physiology

- Experiment at CRL in 2004

  - Tomato, radish and lettuce
  - Measured leaf photosynthetic rates from sunrise to sunset
  - Measured the starch concentrations in tomato leaves during the major growing season
  - Examined the patterns of starch concentration in leaves at night
  - Examined the variation of starch concentrations in leaves and fruit for 24 hours
  - Examined the pattern of starch concentration in leaves from dusk until dawn at Perch Lake

Photosynthesis



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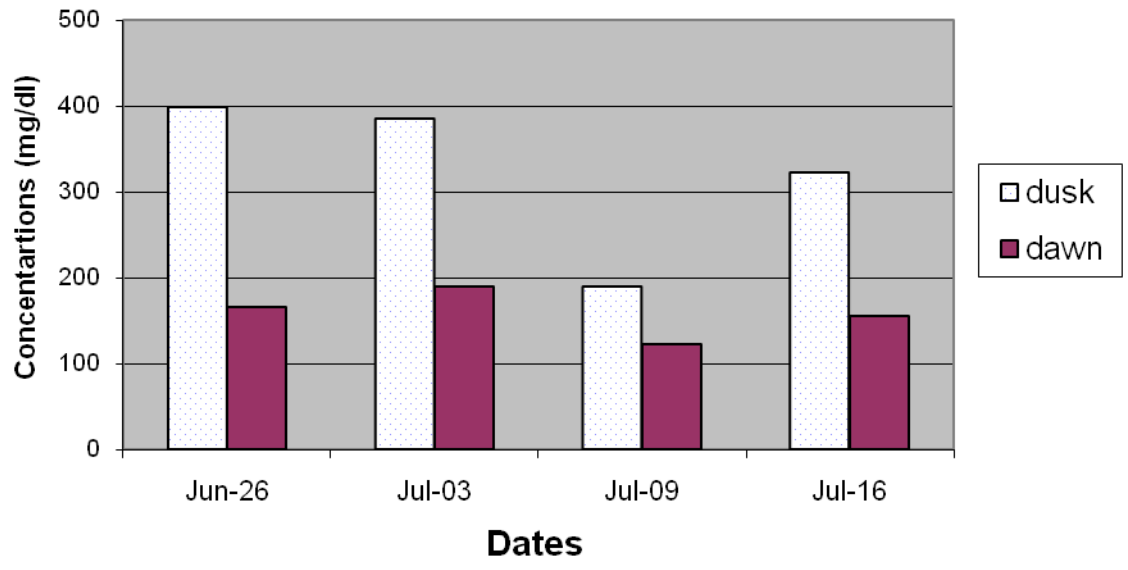
Starch Concentration in Tomato Leaves



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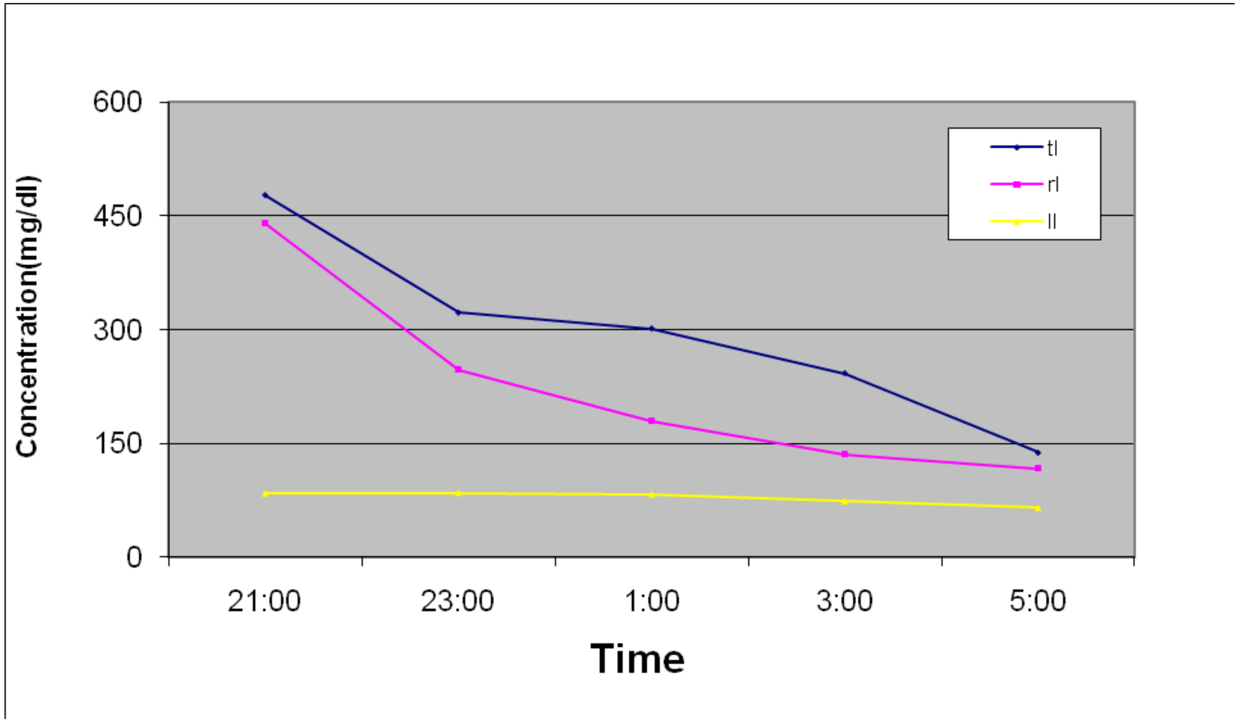
Pattern of Starch Concentration



tl: tomato leave, rl: radish leave, ll: lettuce leaves

OBT Formation Modeling at Night

- Conceptual model

  - Has been developed based on carbohydrate allocation in plants in the dark (2002)

- Mathematical model

  - Has been developed based on a conceptual model of carbohydrate allocation in plants in the dark (2002)

- Implication to ETMOD

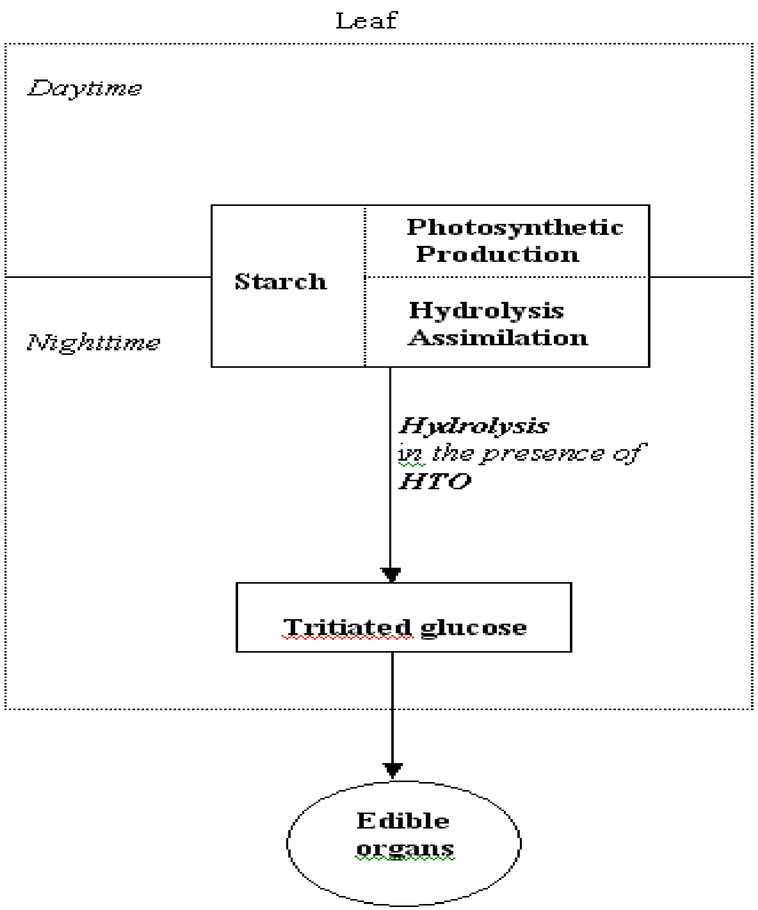
  - The mathematical model will be incorporated into an environmental tritium model to quantify the nocturnal formation of OBT in plants (ongoing)

Conceptual Model

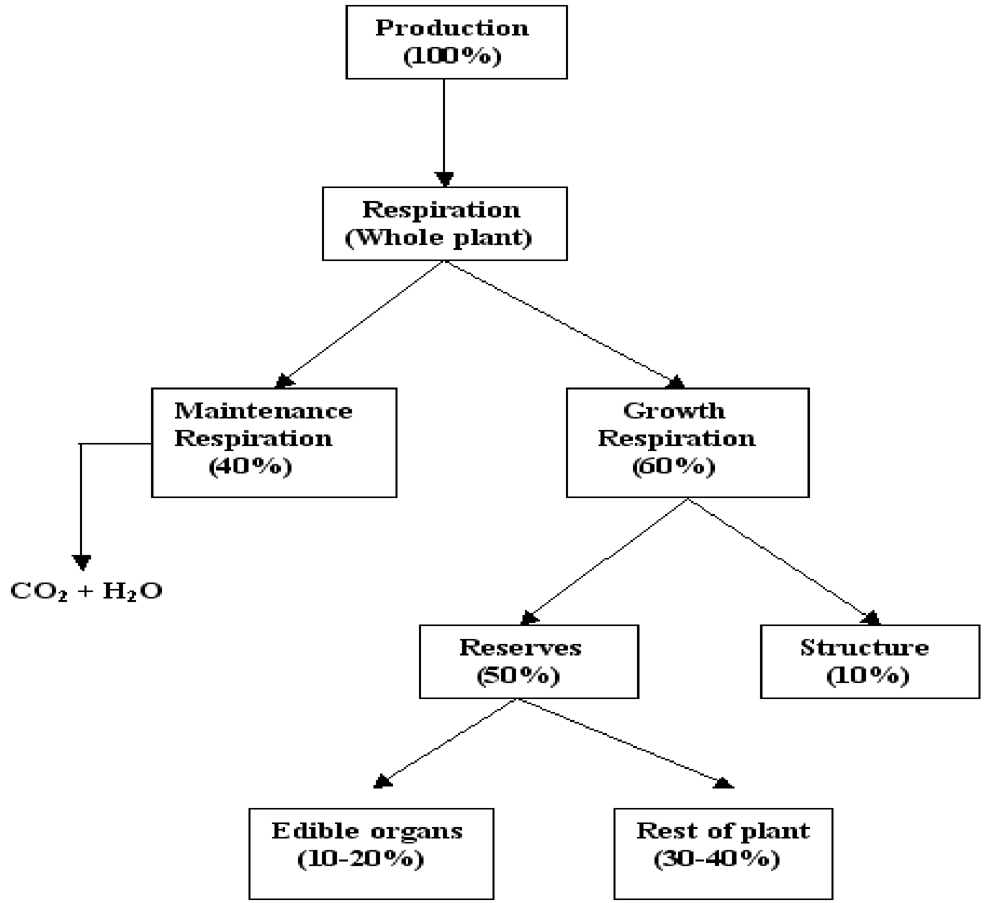
- The conceptual model is composed of two parts
  - the transfer of tritium from air to leaf
  - nocturnal OBT formation

- Assumptions for OBT formation
  - hydrogen will act in concert with carbon in most processes
  - the main processes occurring in plants are starch metabolism and plant growth
  - biological transformation is not considered
  - all the photosynthetic starch produced and stored in the leaves during a given day is to be completely hydrolyzed during the following night
  - HTO is not transferred from leaf to sink
  - there are no processes occurring in the sink at night that result in the incorporation of tritium into organic material

Conceptual Model of OBT Formation



Carbohydrate Allocation in Mature Plant



Mathematical Model (1)

Equation (1)

$$M_w \frac{dC^{\ell}_{TFWT}}{dt} = v_{ex}(C_a - \frac{\rho_s}{\alpha} C^{\ell}_{TFWT})$$

M_w is the mass of plant water per unit area of ground surface (kg m^{-2}),
C^{ℓ}_{TFWT} is the tritium concentration in the leaf water (Bq L^{-1}),
t is time (s),
v_{ex} is the exchange velocity between air and plant (m s^{-1}),
C_a is the tritium concentration in air (Bq m^{-3}),
ρ_s is the density of water vapour in saturated air (kg m^{-3}) and
α = 1.1 is the quotient of T/H ratios in liquid and vapour.

Mathematical Model (2)

Equation (2)

$$C_{OBT}^{f}(\tau) = \frac{0.6}{M_f} A_\ell f_s D\, M_s \frac{\tau}{\tau_n} \overline{C_{TFWT}^{\ell}}$$

M_f is the total fresh weight of all fruit on the plant

A_l is the total leaf area of the plant (dm^2)

f_s is the fraction of hydrolysed starch that is translocated from the leaf to the sink

D is the discrimination factor

M_s is the number of hydrogen atoms

It is probably not worth much because its prediction didn’t agree with observation.

Evaluation of OBT Formation at Night

- The conceptual and mathematical models
- The transfer of HTO from air to leaves (equation 1)
- OBT concentration in edible parts of non-leafy vegetables (equation 2)
- The ratios of predictions to observations range 0.45 to 416, with their geometric mean being 18
- Uncertainties associated with input parameters, observations and deficiencies in the model itself
- How OBT is produced or translated during the day following a night-time tritium exposure

Prediction OBT at Night

Scenario	ETMOD 2 (1996)	ETMOD 2 (2005)	Observation
Nighttime release, HTO in leaves after 2 hours (Bq/mL)	20	500	73,000
Nighttime release, OBT in grain at harvest (Bq/g)	14	6	280
Daytime release, HTO in leaves after 2 hours (Bq/mL)	102,000	102,000	89,000
Daytime release, OBT in grain at harvest (Bq/g)	50	18	140

BIOMOVS II Spring Wheat Scenario

Summary

- **Reinforce the OBT formation theory at night**
  - **- Not much difference between daytime and night time**
- **Model parameters**
  - **- Optimization of various parameters**
- **Validation experiment under various weather conditions**
- **AECL's tritium (HTO and OBT) study**
  - **- Long term project**
  - **- International cooperation**
  - **- Technical difficulties**
  - **- Limited experience and knowledge**

Tritium Guys

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