E FLUX

Easy set-up. Expert results.

MODELING COUPLED HEAT TRANSFER AND HEAT GENERATION: LESSONS FOR MEASURING NSZD RATES USING THERMAL GRADIENT METHODS

JULIO ZIMBRON

2023 BATTELLE CONFERENCE

MAY 9, 2023

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Outline

- NSZD Intro
- Methods
  - The Model: Assembling a (simplified) VCS (Virtual Contaminated Site)
    - Kinetics
    - Mass Balances
    - Heat Balances
  - The Thermal Gradient Method
  - Modeling Coupled Heat Transfer and Generation
  - Another model:
    - "The Single Stick" Method
- Results
- Conclusions

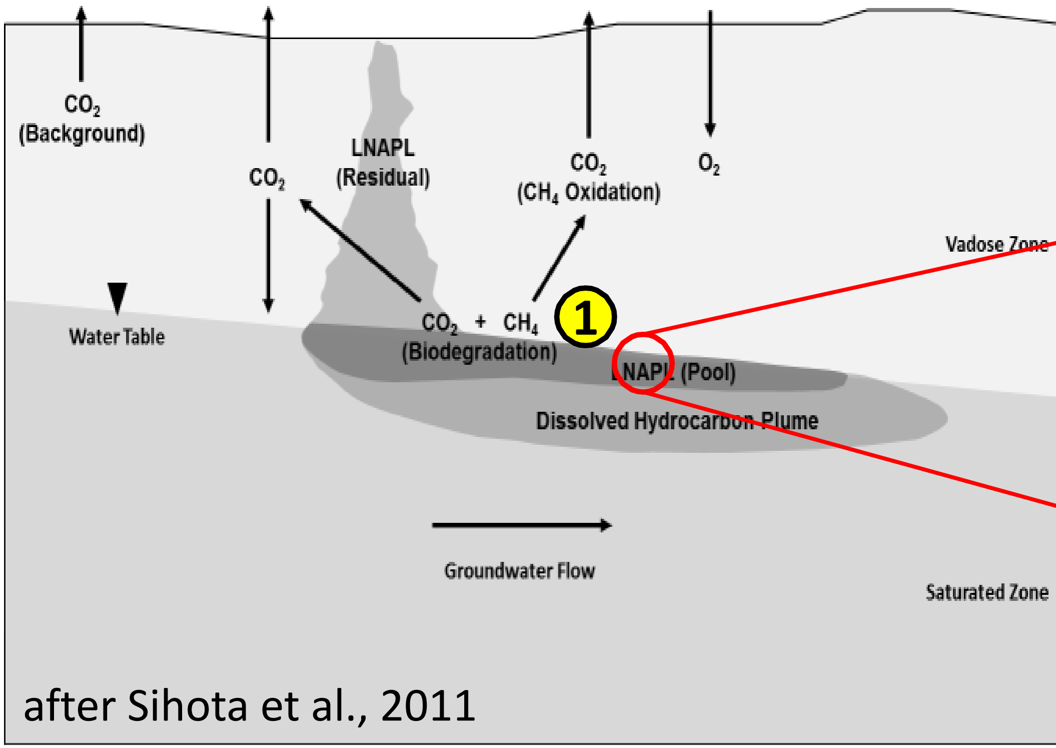
Motivation

- Need to reconcile lab and field data
  - Petroleum biodegradability
  - Biodegradation temperature dependence
- A decision-support tool for contaminated sites
- Specifically: validation of thermal gradient method

http://avecom.be/product/microcosm-tests

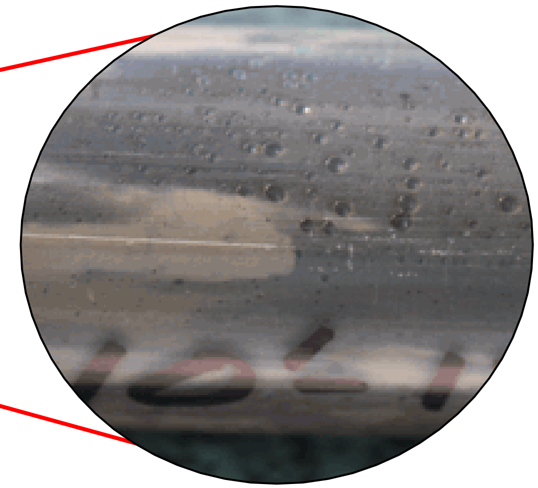
http://www.mprnews.org/story/2014/06/03/bemidji-oil-spill-site-research

NSZD Conceptual Model



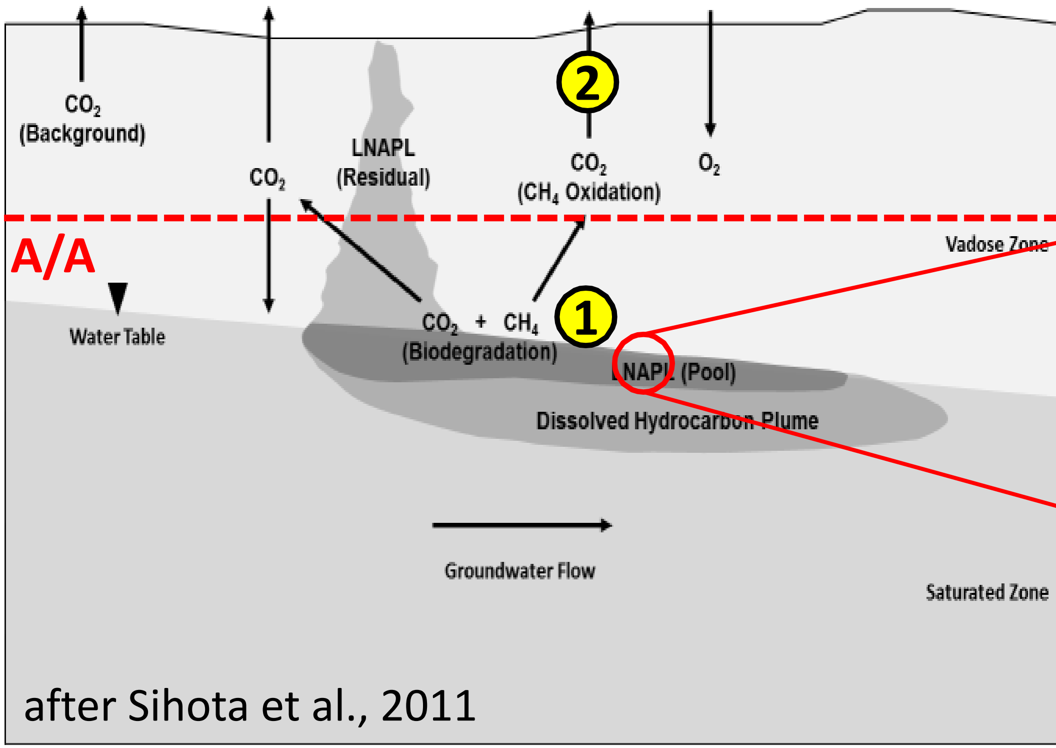
after Sihota et al., 2011

Microbial processes produce stoichiometric amounts of energy



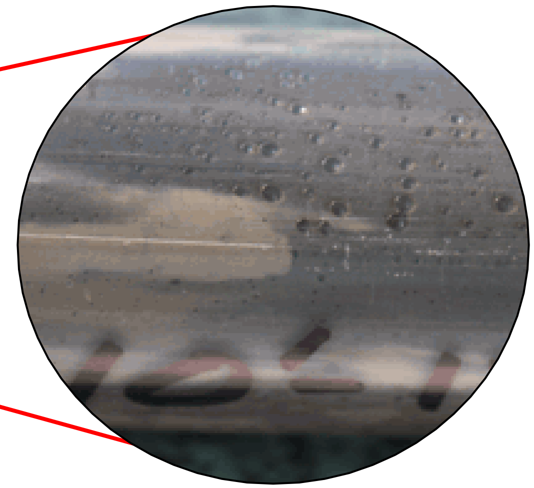
1 $C_8H_{18} + 3.5O_2 \rightarrow 6.25CH_4 + 1.75CO_2 = 24$ kcal/gHC

NSZD Conceptual Model



after Sihota et al., 2011

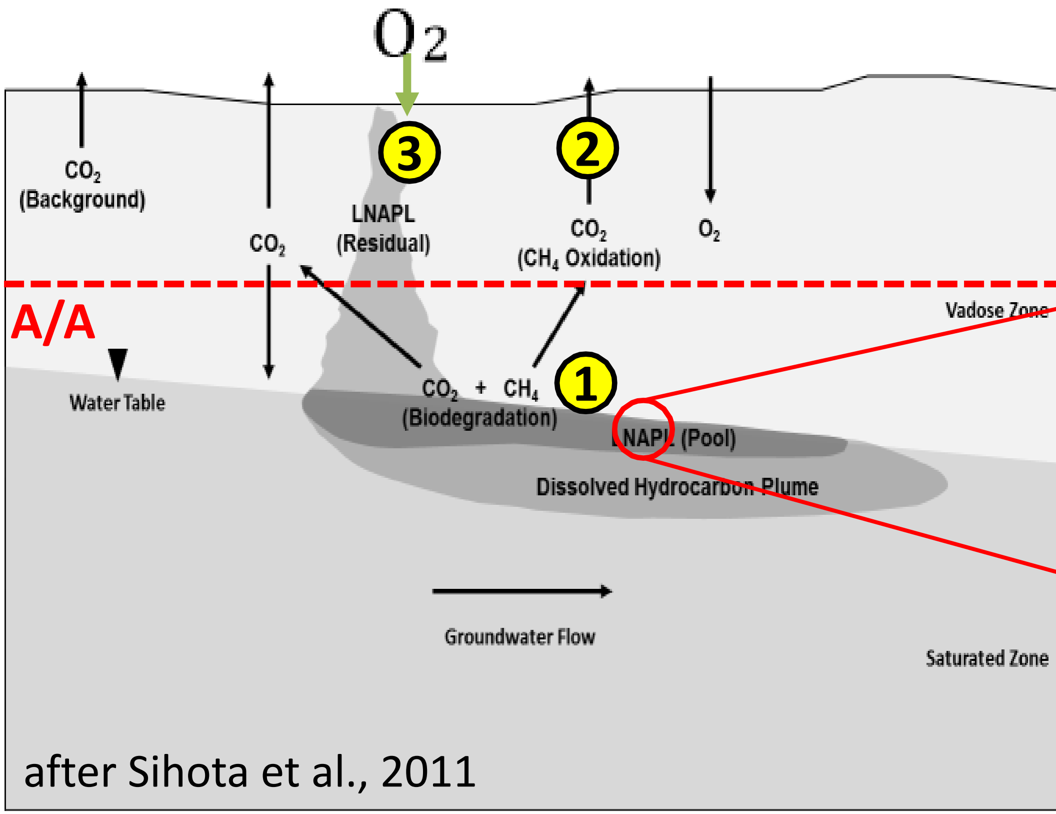
Microbial processes produce stoichiometric amounts of energy



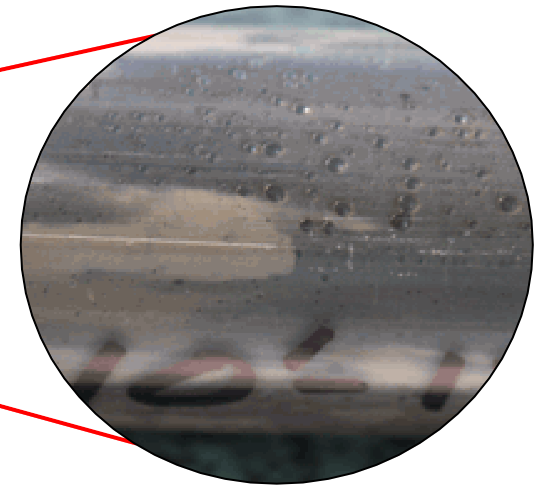
1 $C_8H_{18} + 3.5O_2 \rightarrow 6.25CH_4 + 1.75CO_2 \quad \Delta H = 24\ \text{kcal/gHC}$

2 $6.25\ CH_4 + 12.5O_2 \rightarrow 6.25CO_2 + 12.5H_2O \quad \Delta H_{MOx} = 1{,}200\ \text{kcal/gH}$

NSZD Conceptual Model

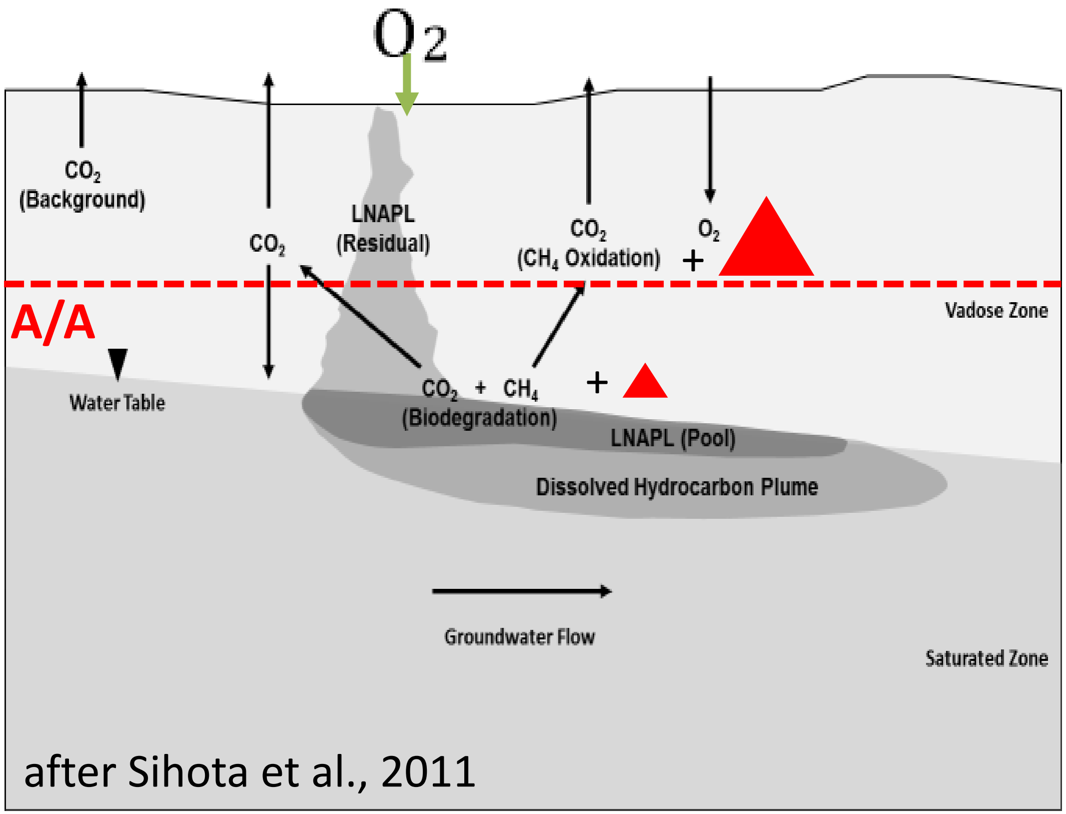


Microbial processes produce stoichiometric amounts of energy



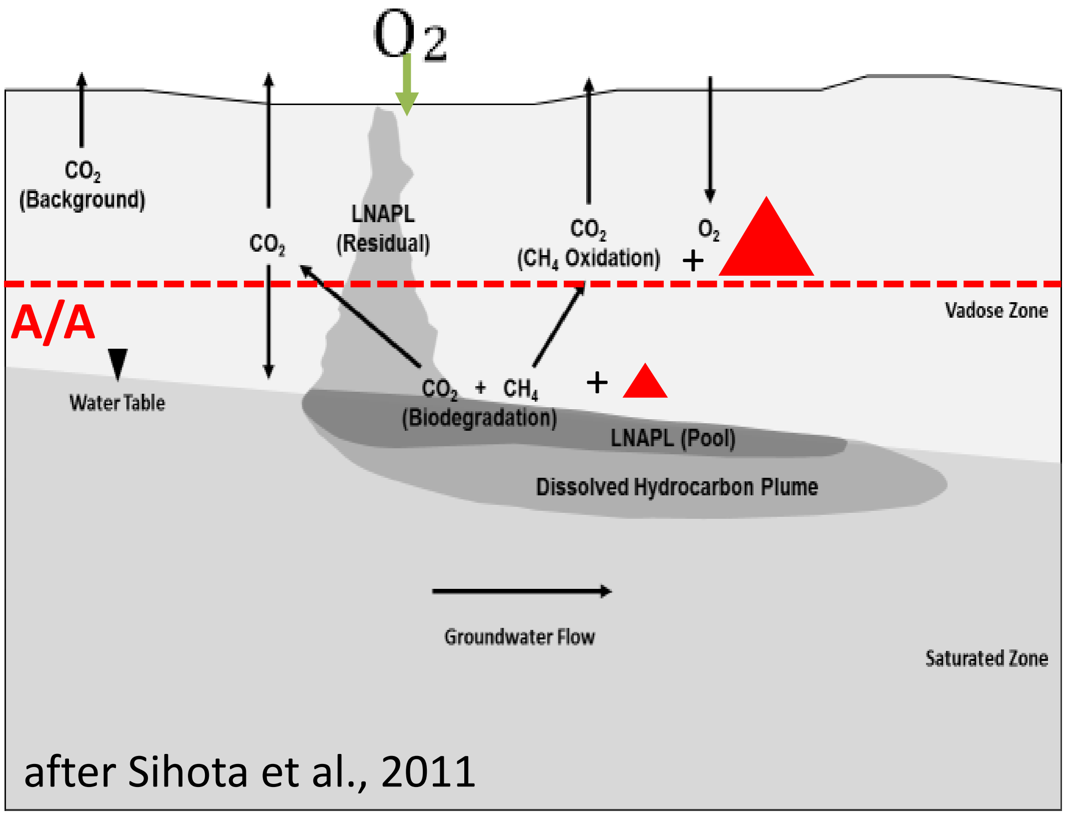
1. $C_8H_{18} + 3.5O_2 \rightarrow 6.25CH_4 + 1.75CO_2 \quad \Delta H_{CH_4} = 24\ kcal/gHC$
2. $6.25\ CH_4 + 12.5O_2 \rightarrow 6.25CO_2 + 12.5H_2O \quad \Delta H_{MOx} = 1,200\ kcal/gH$
3. $C_8H_{18} + 12.5O_2 \rightarrow 8CO_2 + 9H_2O \quad \Delta H_{aerobic} = 1,224\ kcal/g$

NSZD methods



after Sihota et al., 2011

NSZD methods



Reaction By-Product	Basis	Implementation
Chemical products	Mass balance	CO_2 flux measurements
Heat	Heat balance	Heat flux measurements

Modeling Biodegradation Kinetics

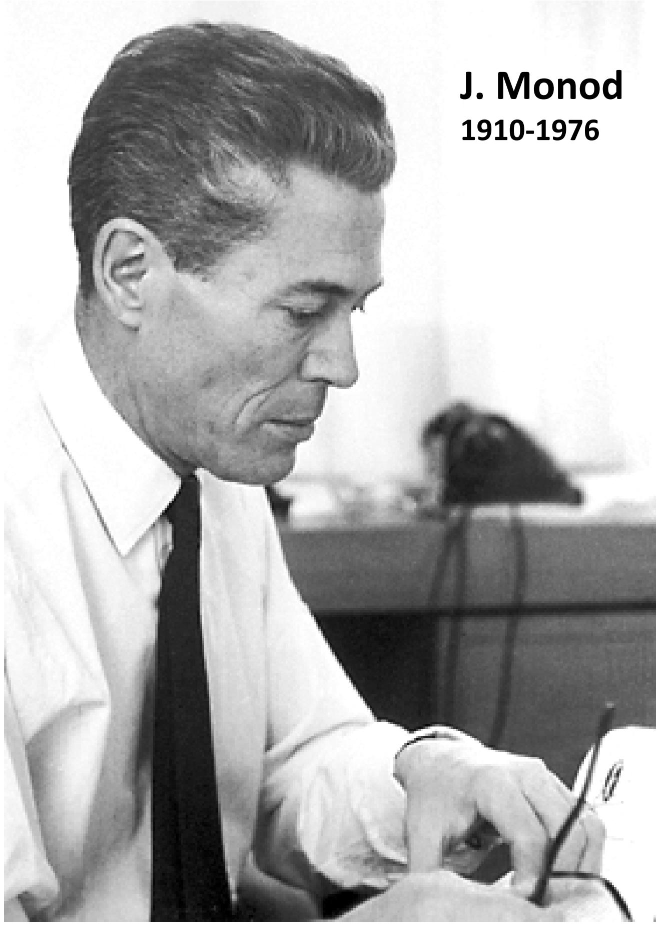
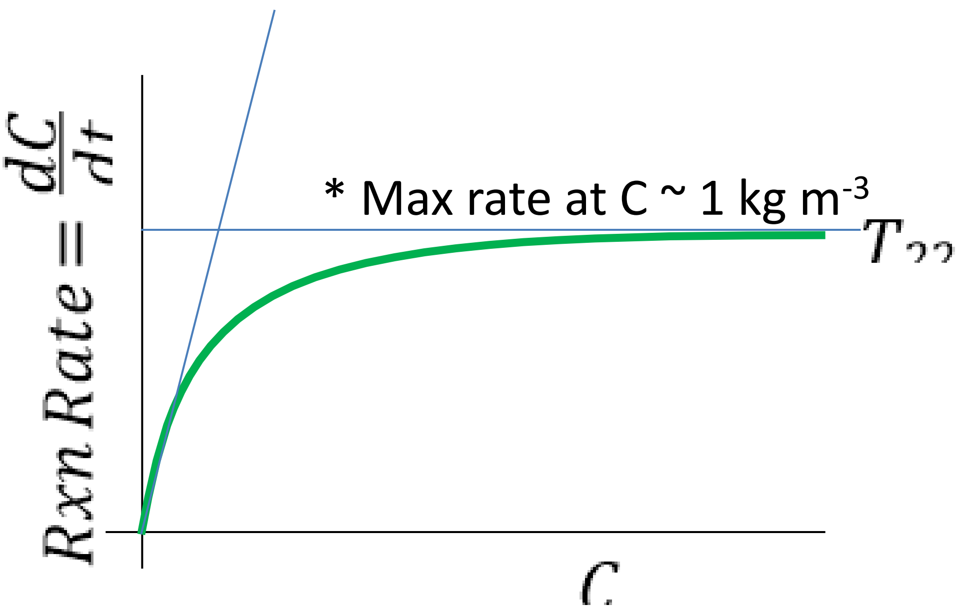


Image from Wikipedia

Reaction Rates (Lab)

Monod Kinetics: Reaction rates depend on **Contaminant Concentration**

$$-\frac{dC}{dt} = \frac{k_{max}C}{C + C_m}$$



1. $C_8H_{18} \rightarrow CH_4$
   - Siddique et al. (2008)
   - k_{max} = 1.54 kg m^{-3} yr^{-1}
   - C_m = 0.47 kg m^{-3}
2. $CH_4 \rightarrow CO_2$
   - Fast reaction (Davis, 2009)
3. $C_8H_{18} \rightarrow CO_2$
   - Aerobic rates higher than anaerobic
   - (Molins et al., 2010)

* Compare 1 kg/m3 = 1 g/L to

Sr = 0.1 (smallest value from Mercer and Cohen, 1990)

-> ~40 g/L so most sites with NAPL likely operate at Max rate (0 order)

Modeling (Temperature-Dependent) Biodegradation Kinetics

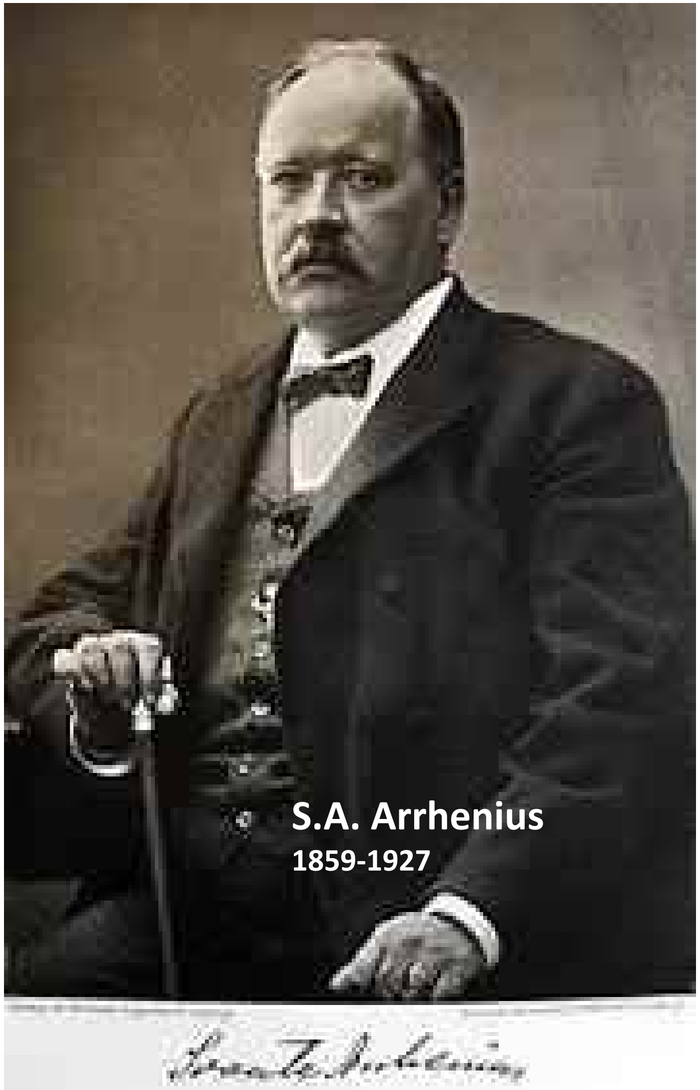
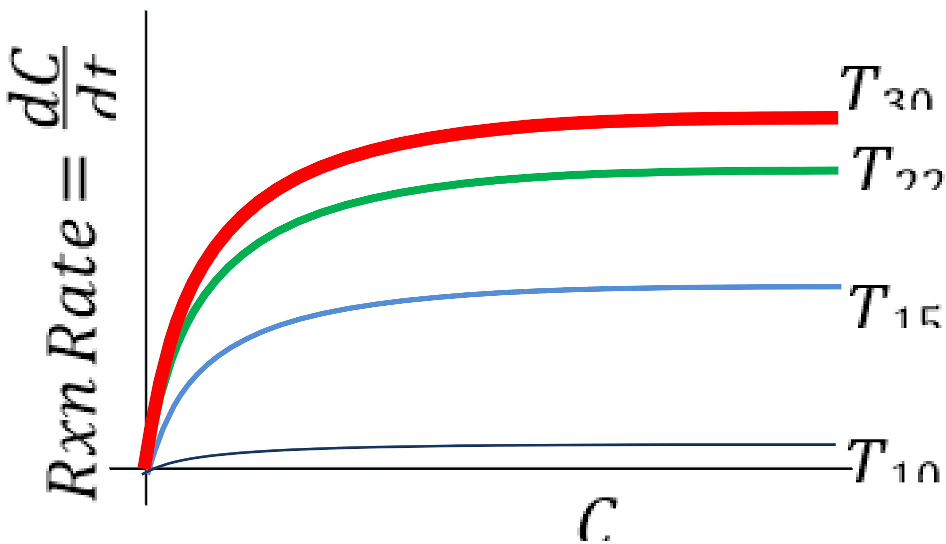


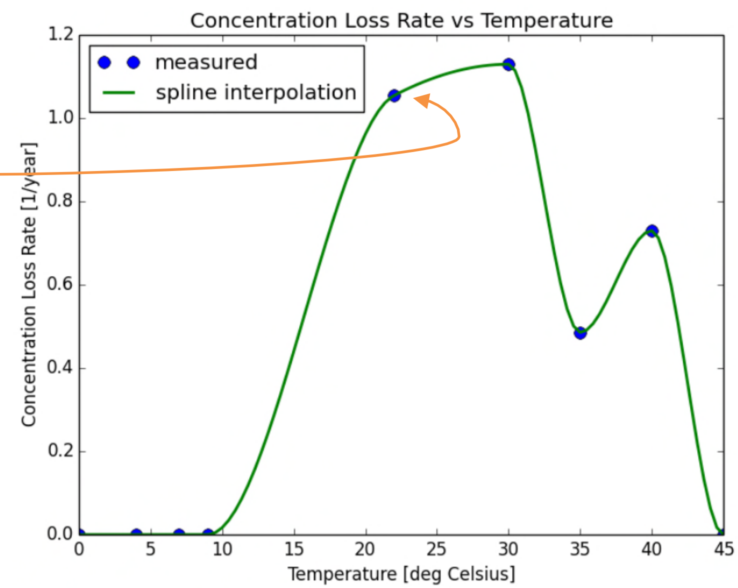
Image from Wikipedia

Reaction Rates (Lab)

Reaction rates depend on **Temperature** (Arrhenius equation)

$$k = Ae^{\frac{-E_A}{RT}}$$





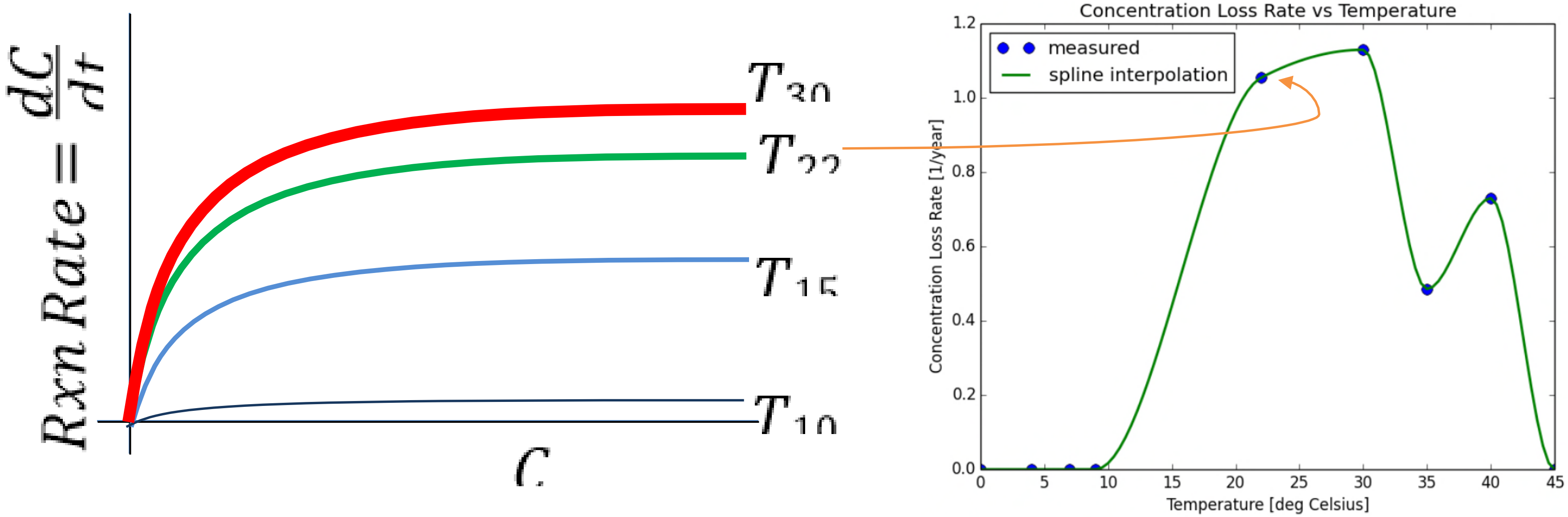
Data from

Zeman, N. et al, 2014

Reaction Rates (Lab)

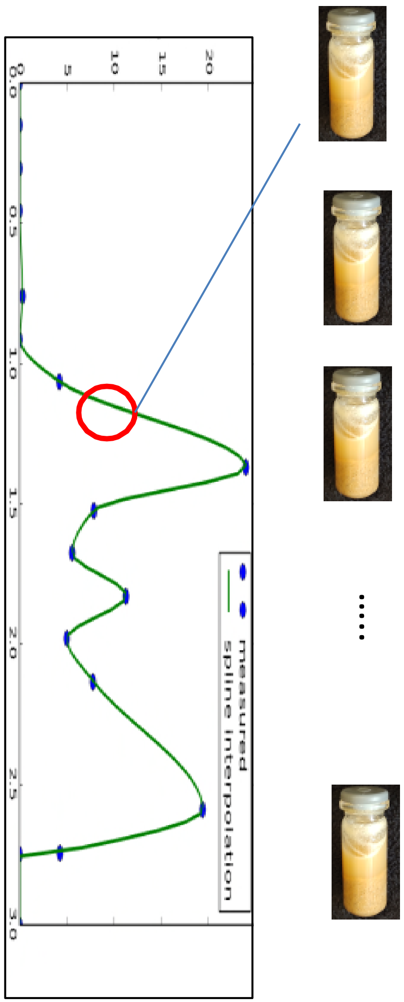
Reaction rates depend on **Temperature** (Arrhenius equation)

$$k = Ae^{\frac{-E_A}{RT}}$$



Model Approach

Inputs



Approach

At each elevation account for

a) Local LNAPL concentration

b) Correct for local temperature

c) Estimate "local biodegradation rate"

d) Cumulative biodegradation rate results in a bulk methane oxidation rate at A/A interface

Last Piece: Modeling Heat Transfer in Soils

Heat Equation with Heat Generation

To solve, need

- Soil properties (density, heat capacity and heat transmissivity)
- Boundary conditions (i.e., ambient and groundwater temperature)

Heat equation

$$+ qi \times \frac{1}{\rho C_p}$$

+ heat generation

Heat Equation with Heat Generation

To solve, need

- Soil properties (density, heat capacity and heat transmissivity)
- Boundary conditions (i.e., ambient and groundwater temperature)

Heat equation

$$+ qi \times \frac{1}{\rho C_p}$$

+ heat generation

q_i : heat generation rate

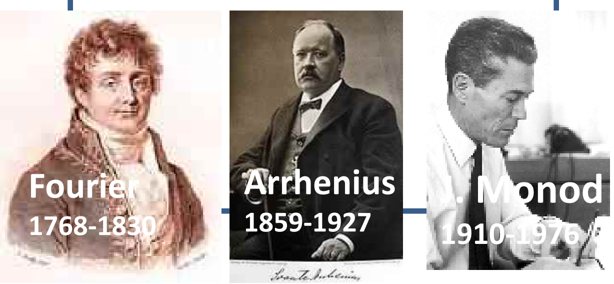
1. $C_8H_{18} \rightarrow CH_4$
2. $CH_4 \rightarrow CO_2$
3. $C_8H_{18} \rightarrow CO_2$

Heat generation rate stoichiometric to the reaction rate

Modeling Heat Transfer in Soils

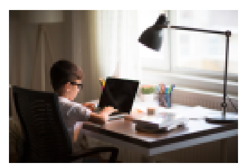
Field Data

Laws of Nature



A simple, yet realistic geometry

Model
(Heat Equation)



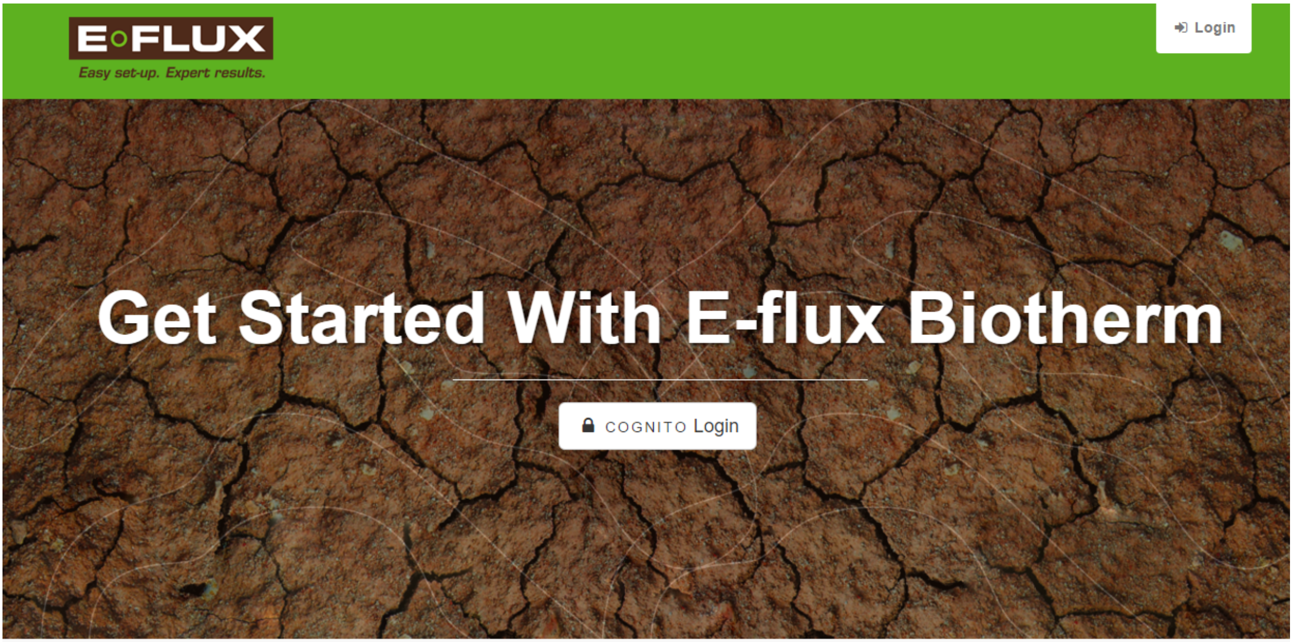
Outputs:

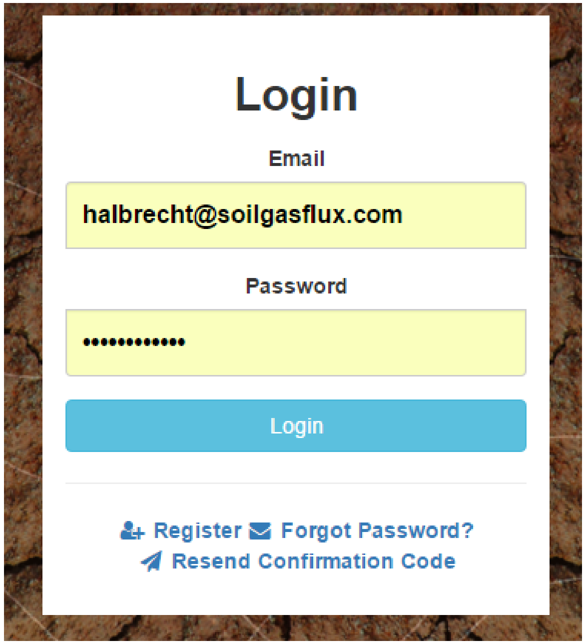
$$T_1(t,z)$$
$$C_1(t,z)$$
$$NSZD\ rate = \sum_z \frac{\Delta C}{\Delta t}$$

Formerly available at:

www.BiogenicHeat.com

Web Based Model, Open to Anyone





To log in, input the info printed at the top of the page

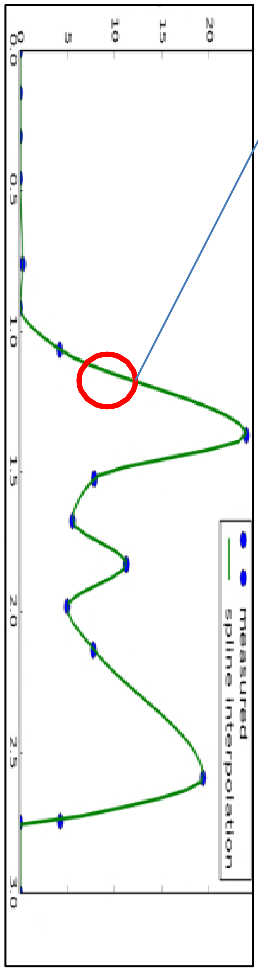
Loging In:

www.soilgasflux.com

www.BiogenicHeat.com

Model Approach

Inputs



Approach

At each elevation account for

a) Local LNAPL concentration

b) Correct for local temperature

c) Estimate “local biodegradation rate”

d) Cumulative biodegradation rate results in a bulk methane oxidation rate at A/A interface

Solve coupled

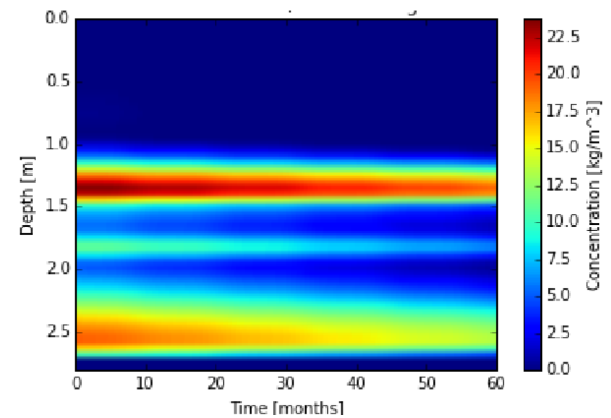
Local temperatures determined by

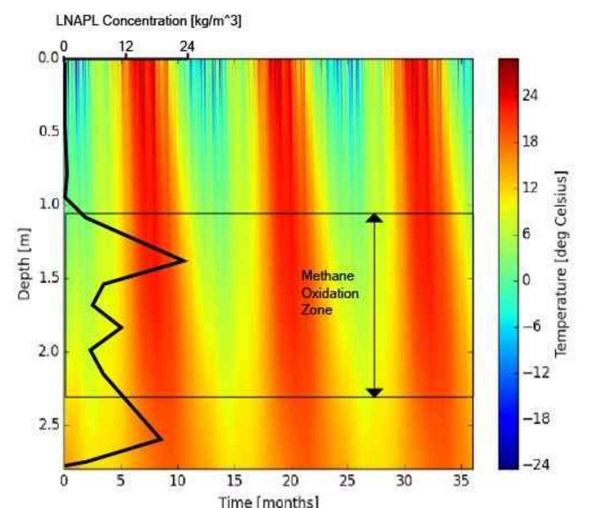
a) Boundary conditions

b) Heat produced by reactions

c) Soil heat transfer

Outputs



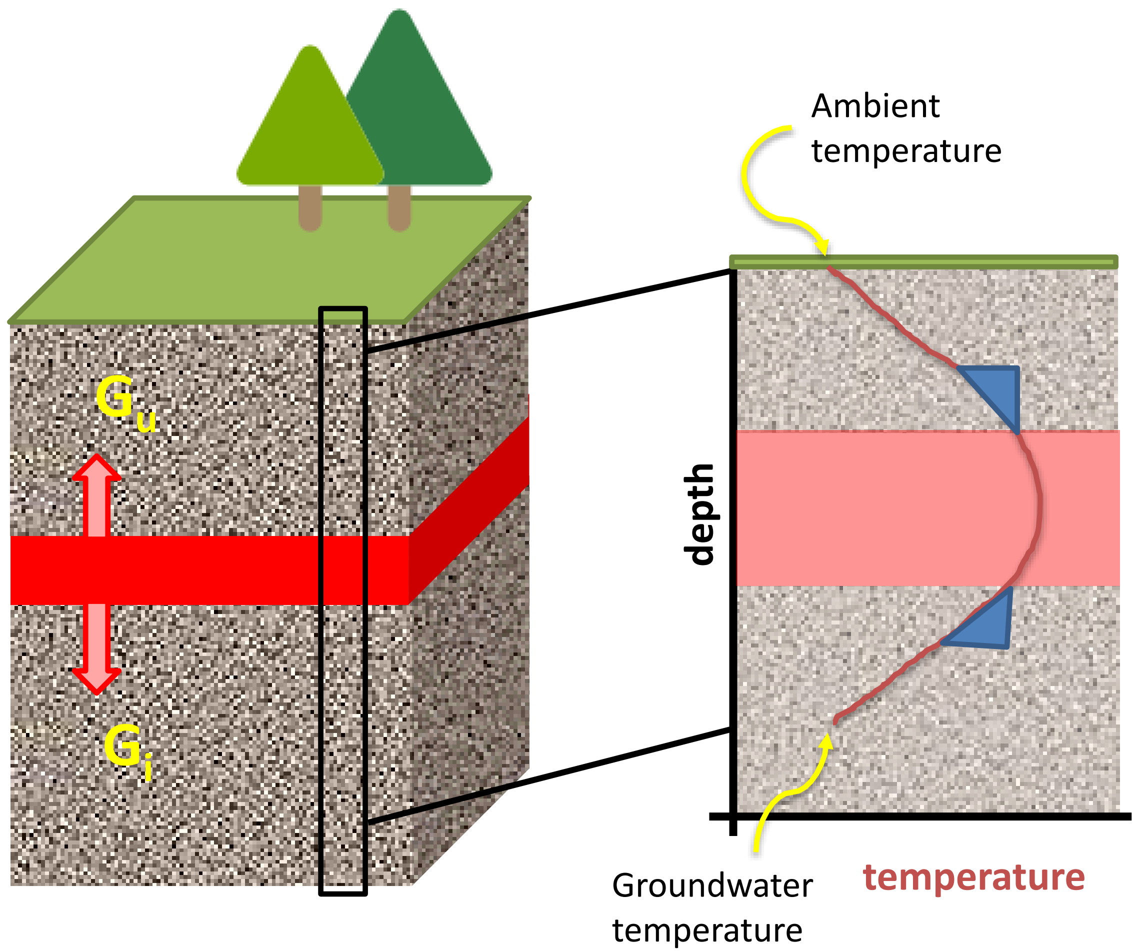


Estimating LNAPL Loss from Heat Balance

Using thermal gradients

$$G = G_u + G_i$$

$$R_{NSZD} = \frac{G_{NSZD}}{\Delta H_{NSZD}}$$

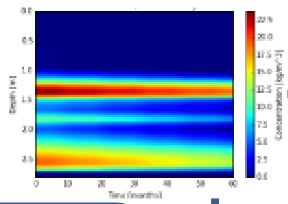


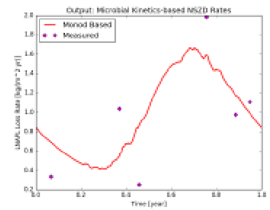
Model Motivation

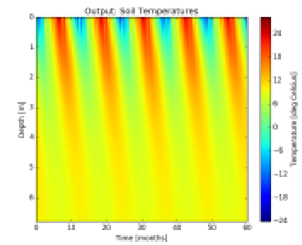
- How NSZD rates vary seasonally?
- How much supplementary heat need to increase NSZD rates (thermally enhanced NSZD)
- Can NSZD rates from the mass balance (i.e., Monod) with those from thermal gradients be reconciled?
    - Background correction
    - Temporal effects (noise)

Two Methods for Estimating LNAPL Loss

Using Mass Balance





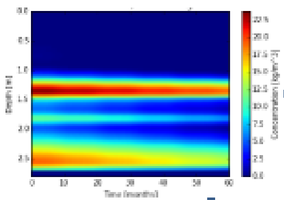


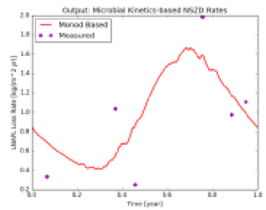
Inputs (Loc. 1) → Model (Heat equation) →

$$T_1(t,z)$$
$$C_1(t,z)$$
$$\mathrm{NSZD}_{Monod} = \sum_z \frac{\Delta C}{\Delta t}$$

Two Methods for Estimating LNAPL Loss

Using Mass Balance





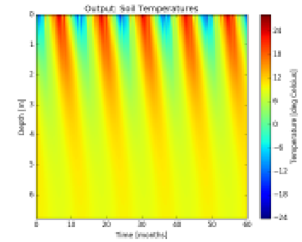
Inputs (Loc. 1) → Model (Heat equation) →

$T_1(t,z)$

$C_1(t,z)$

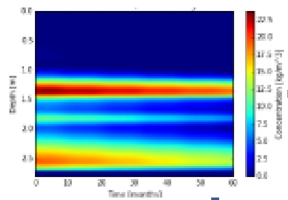
$$\mathrm{NSZD}_{Monod} = \sum_{z} \frac{\Delta C}{\Delta t}$$

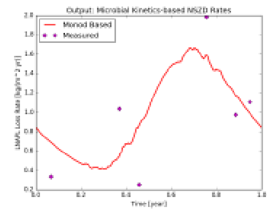
Using Thermal Gradients



Two Methods for Estimating LNAPL Loss

1. Using Mass Balance

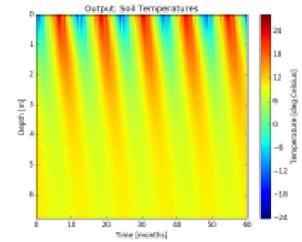


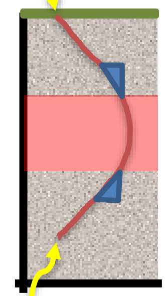


Inputs (Loc. 1) → Model (Heat equation) →

$$T_1(t,z)$$
$$C_1(t,z)$$
$$\mathrm{NSZD}_{Monod} = \sum_z \frac{\Delta C}{\Delta t}$$

2. Using Thermal Gradients





→ **Thermal Gradients**

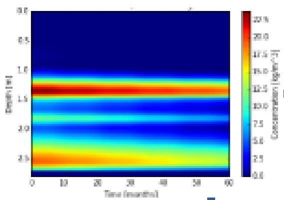
$$\frac{dT}{dz}$$

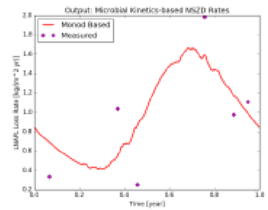
→ **Thermal Gradient-based NSZD rate**

$$R_{\mathrm{TG},loc} = \sum_z \frac{\Delta C}{\Delta t}$$

Two Methods for Estimating LNAPL Loss

1. Using Mass Balance





Inputs (Loc. 1) → Model (Heat equation) →

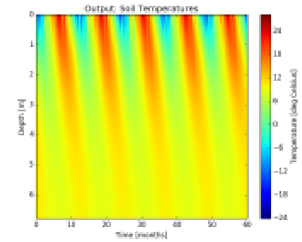
$T_1(t,z)$

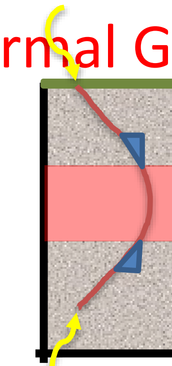
$C_1(t,z)$

$$\mathrm{NSZD}_{Monod} = \sum_z \frac{\Delta C}{\Delta t}$$

Inputs (Loc. 2) → Model (Heat equation) → **Outputs:** $T_2(t,z)$

2. Using Thermal Gradients





Thermal Gradients

$$\frac{dT}{dz}$$

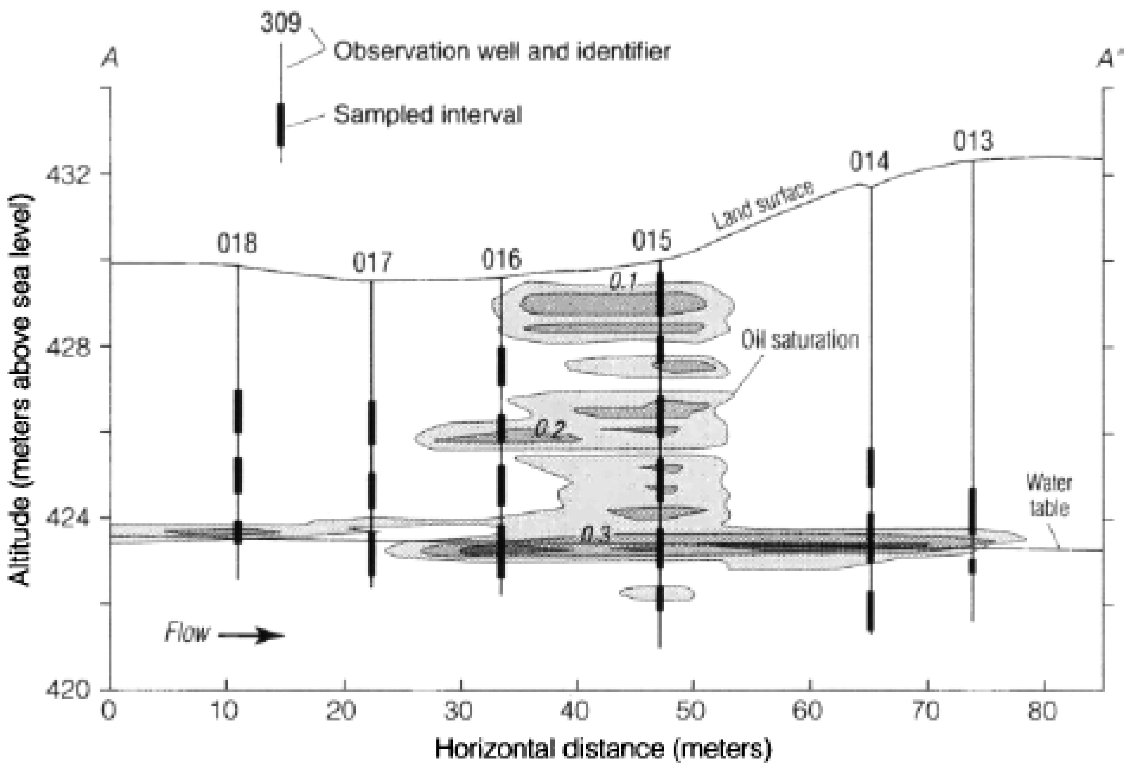
→

Thermal Gradient-based NSZD rate

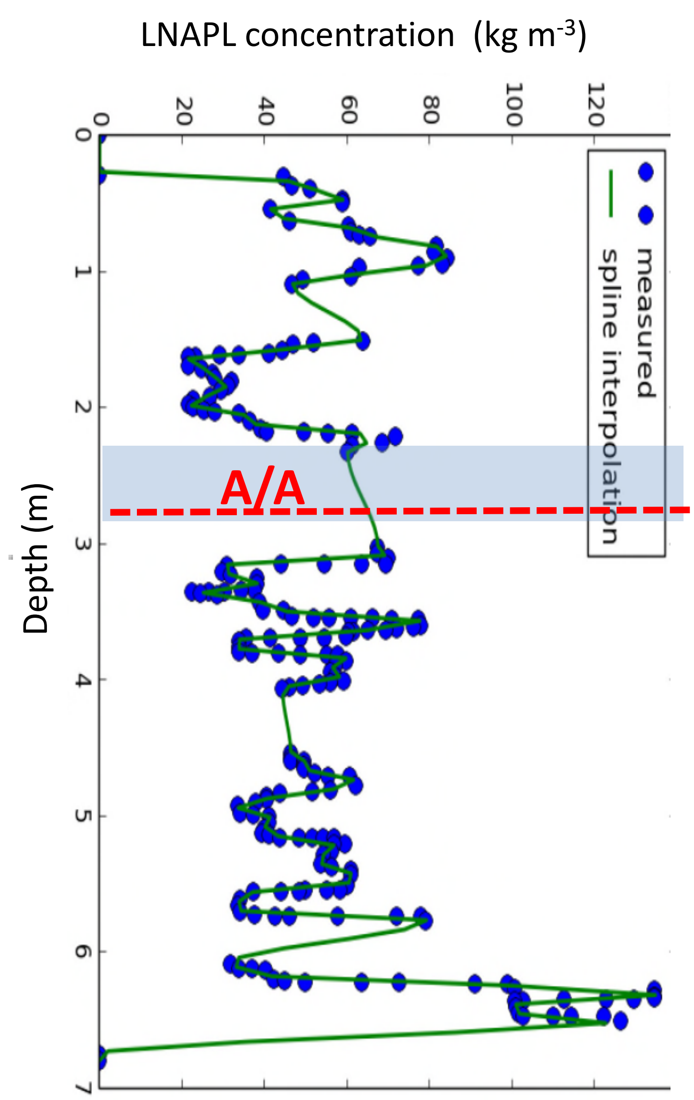
$$R_{\mathrm{TG},loc} = \sum_z \frac{\Delta C}{\Delta t}$$

Case 1: Bemidji

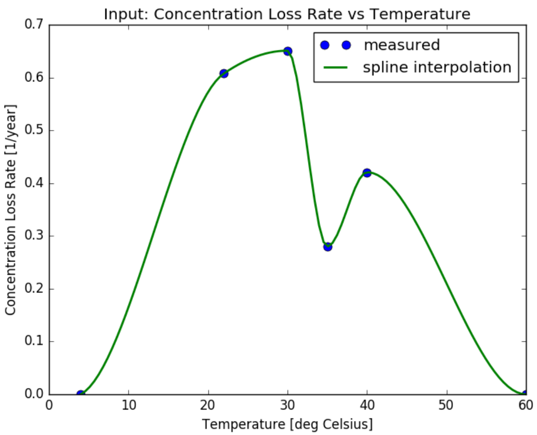
- Crude oil spill site
- Depth to Groundwater: 7 m
- Average Groundwater Temperature: 9 °C

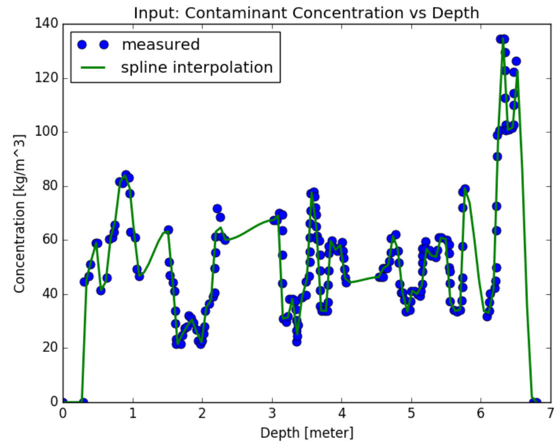


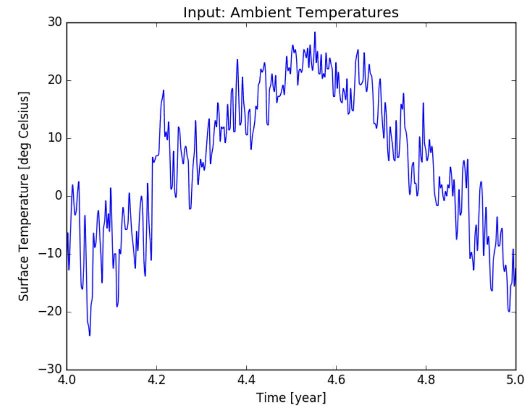
(Dillard et al., 1997)

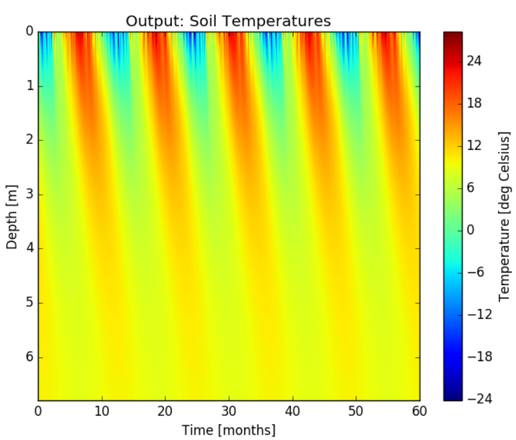


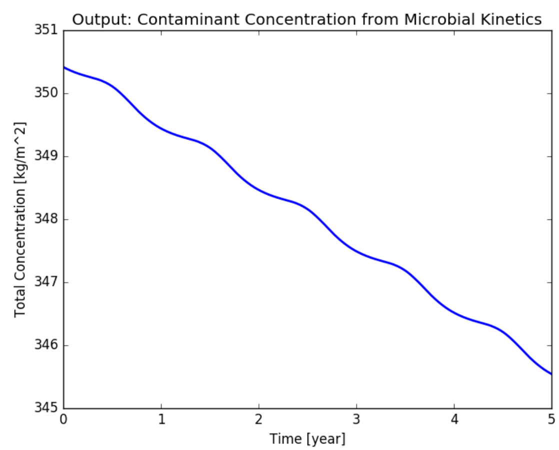
Base Case : Bemidji

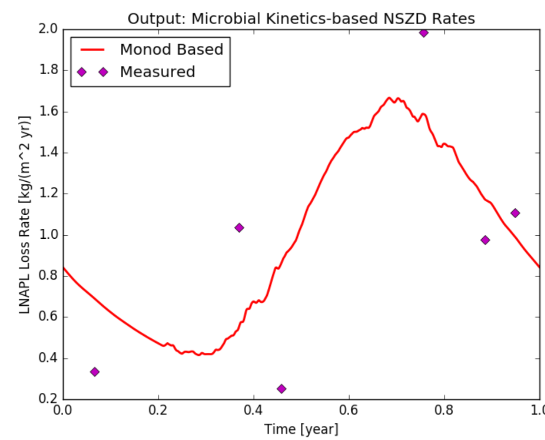








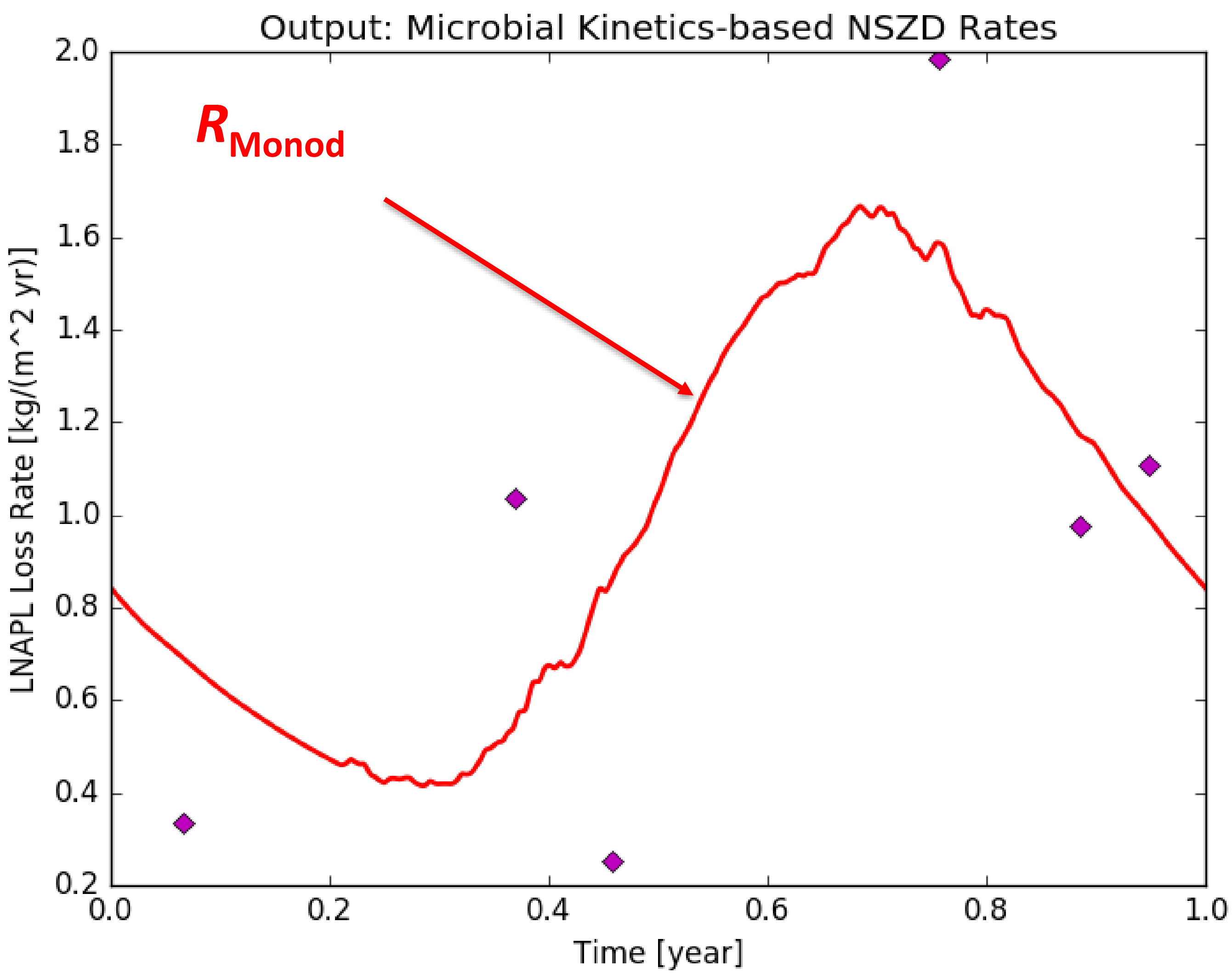




Field rates from Sihota, 2014.

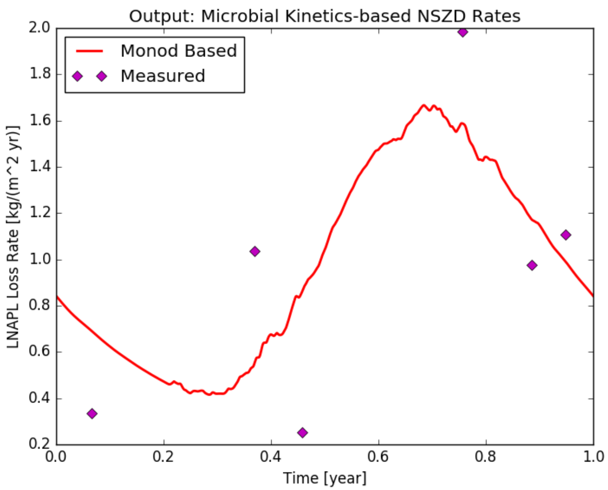
Case Study: Bemidji

Also looking at each set of results in different time scales (short term, monthly, and annual averages) and comparing to the mass-balance NSZD rate (R_{monod})



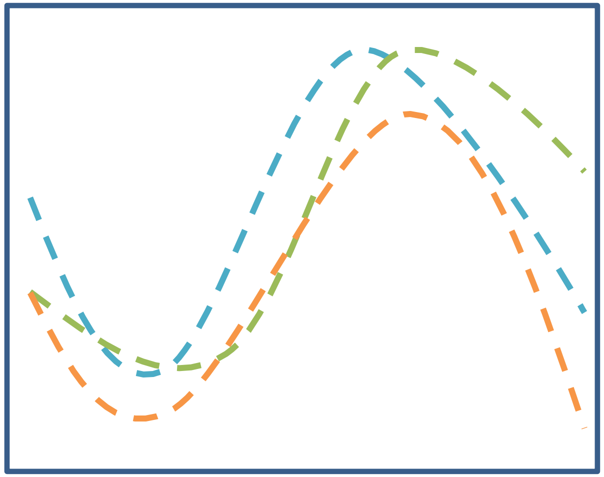
Reconciling MB with HB, V.0?

1. Using the mass balance/ Monod rates



Vs.

2. Using thermal gradients

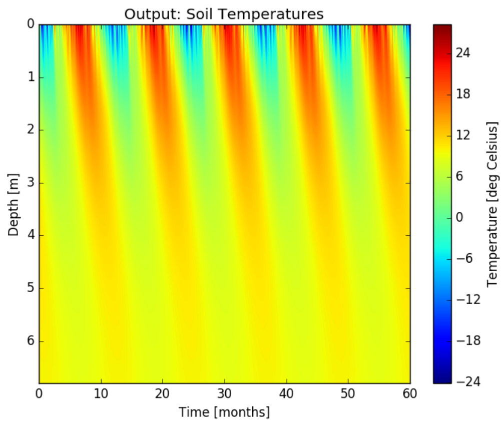


No Background Correction

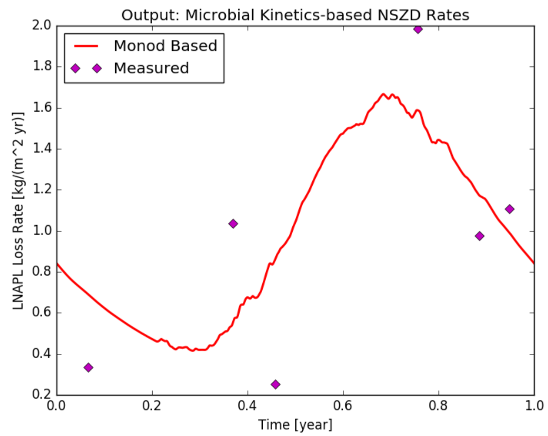
$\alpha_{site} = 3.58 \times 10^{-07}\ m^2/s$

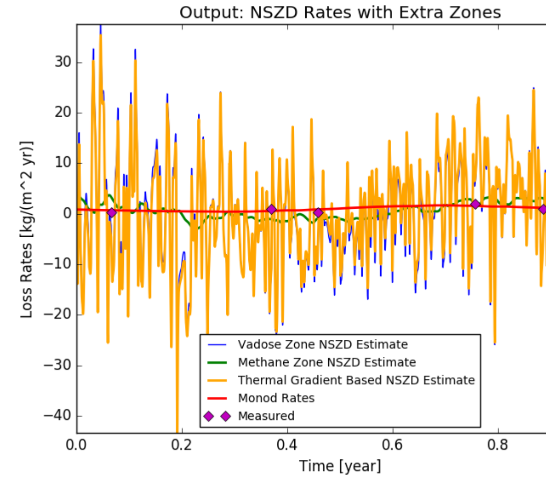
1

Model Output

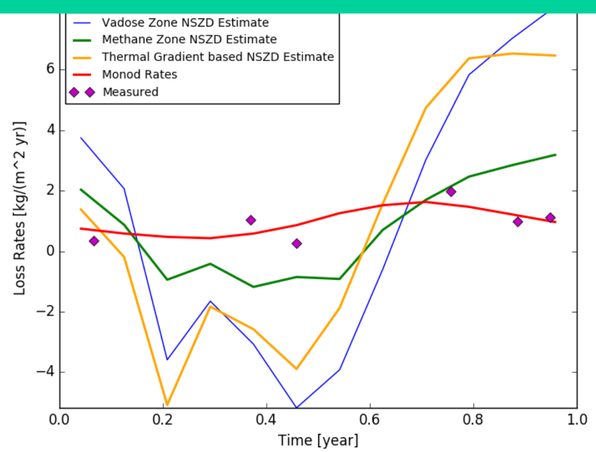


Short term Average Thermal Gradient NSZD rates





Monthly Average Thermal Gradient NSZD rates



Annual Average Thermal Gradient NSZD rates

1. Thermal gradient location	Error Rate
Methane oxidation zone	**26.78%**
Aerobic Zone	**0.64%**
Entire Vadose Zone	**-0.57%**

Model Runs

1

Parameters: Bemidji
Background location: None

2
7

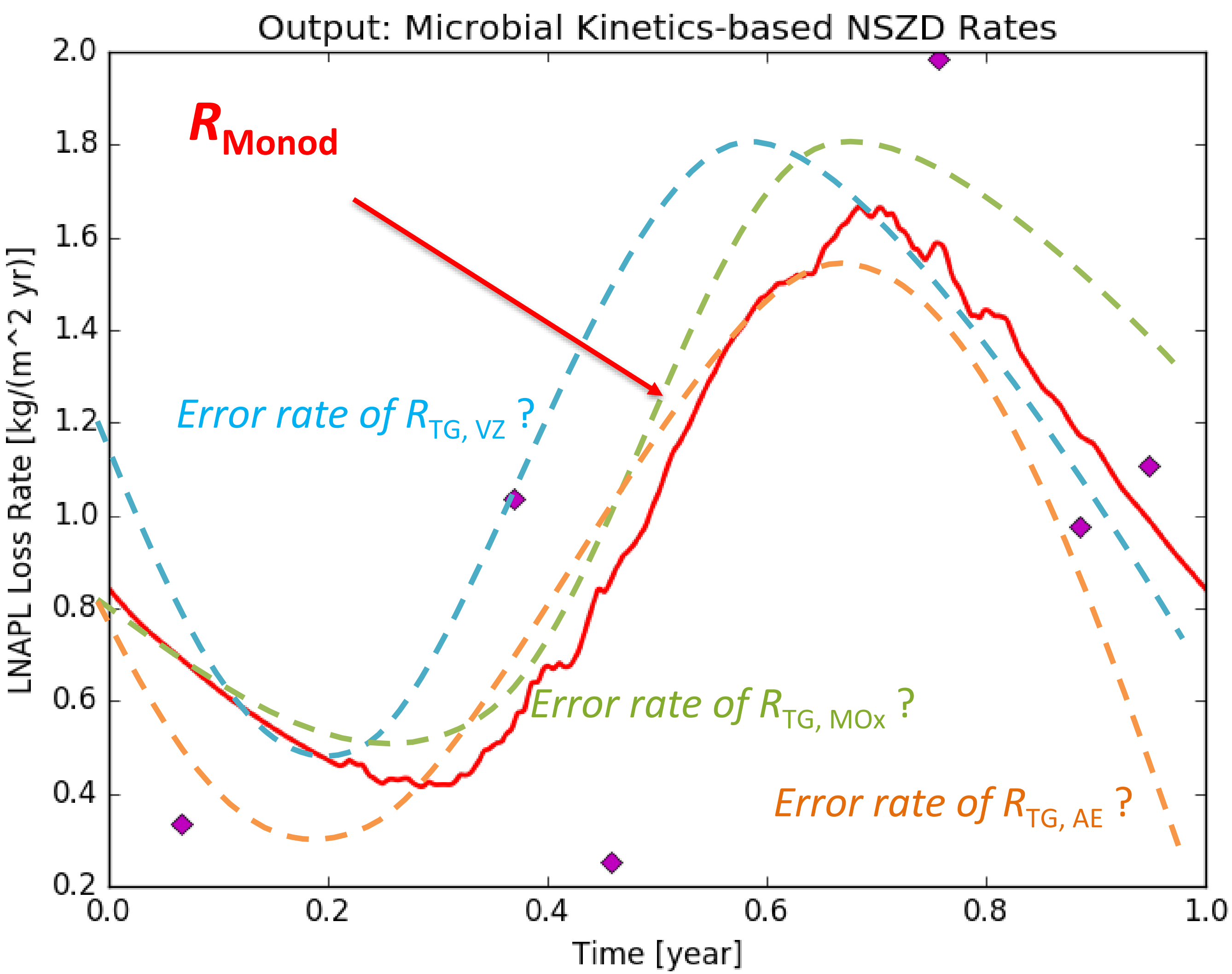
Parameters: Bemidji
Background location: Identical, except no contaminant

3

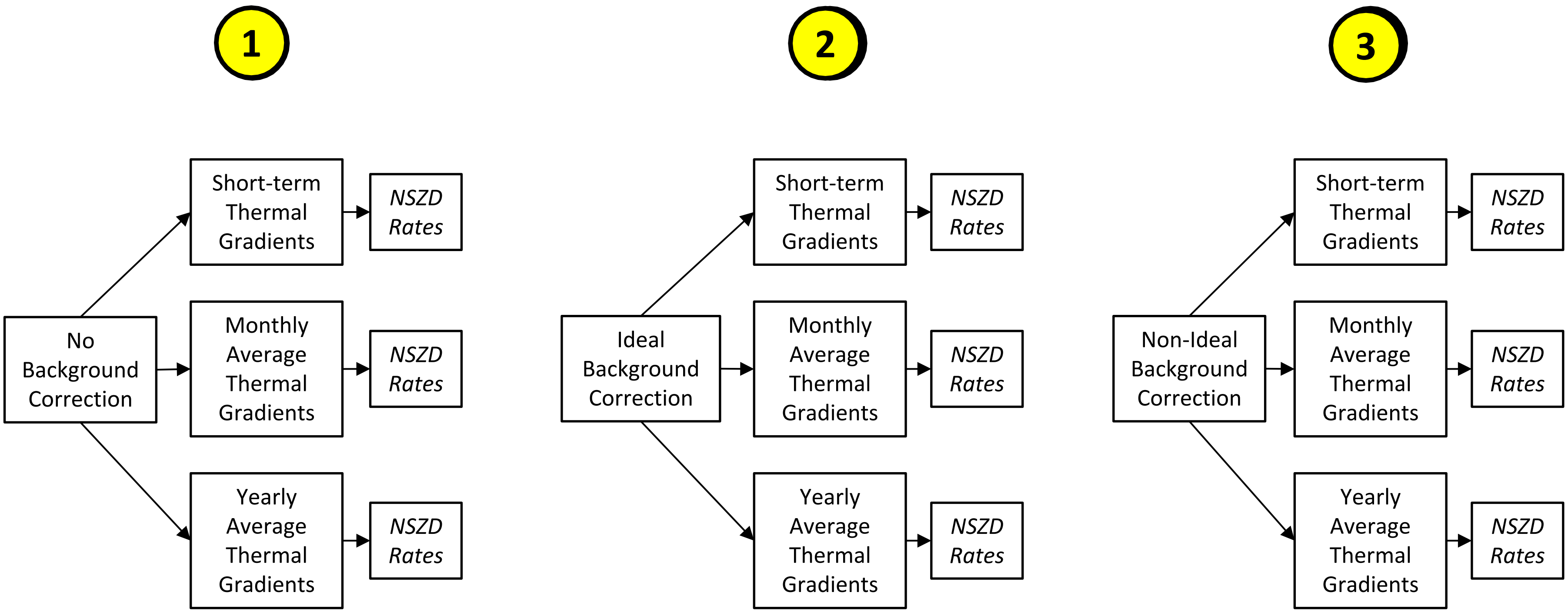
Parameters: Bemidji
Background location: Identical, except different /thermal diffusivity (2x)

Case Study: Bemidji

Also looking at each set of results in different time scales (short term, monthly, and annual averages) and comparing to the mass-balance NSZD rate (R_{monod})



Experimental Design



3 pairs of planes for heat balance

- ***A**erobic zone* ($R_{TG,\ \mathbf{AE}}$)
- *Entire **v**adose **z**one* ($R_{TG,\ \mathbf{VZ}}$)
- ***M**ethane **ox**idation zone* ($R_{TG,\ \mathbf{Mox}}$)

27 thermal gradient estimates

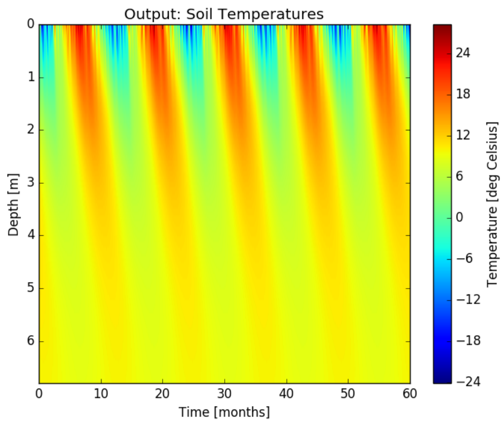
Compare to mass-balance NSZD rates (Monod)

1

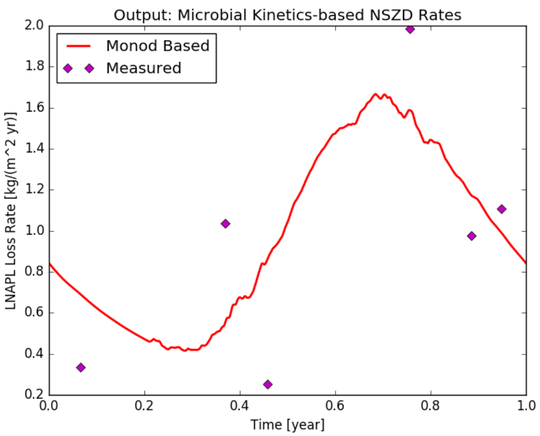
No Background Correction

α_{site} = 3.58 x 10 $^{-07}$ m^2/s

Model Output



Short term Average Thermal Gradient NSZD rates



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Model Output

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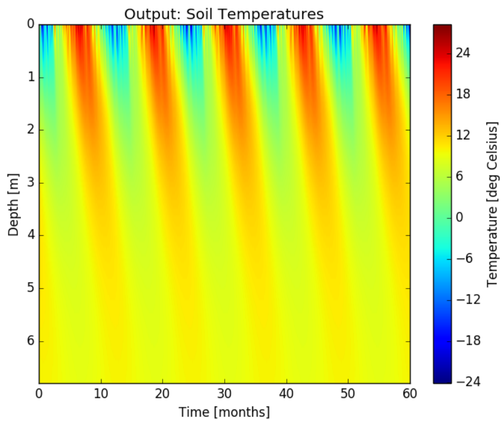

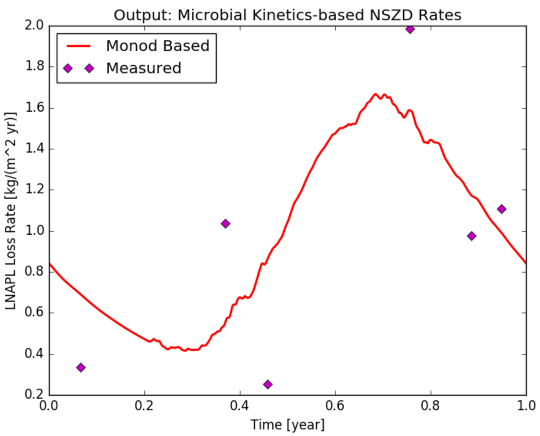

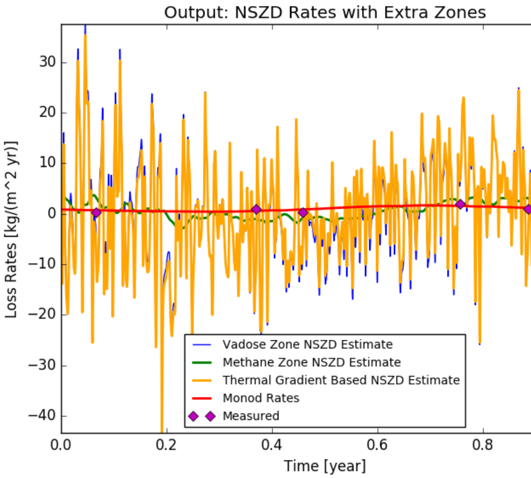

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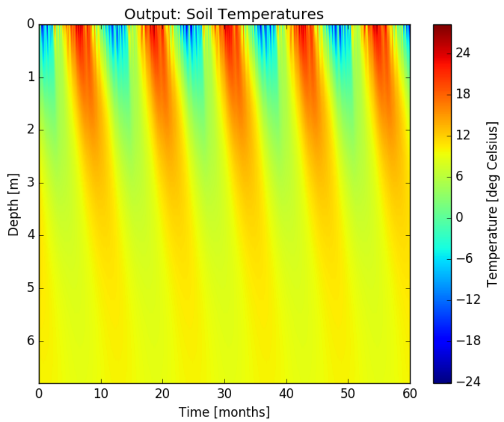
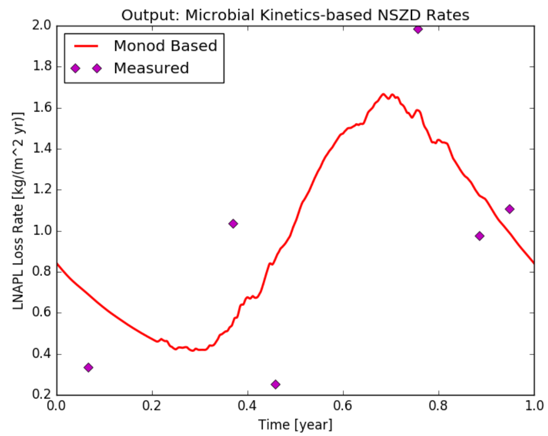
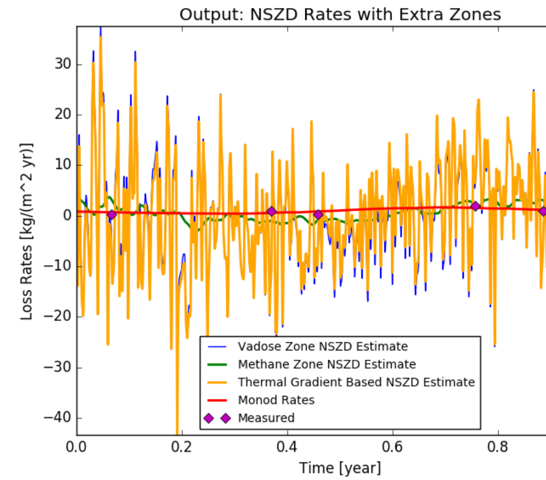
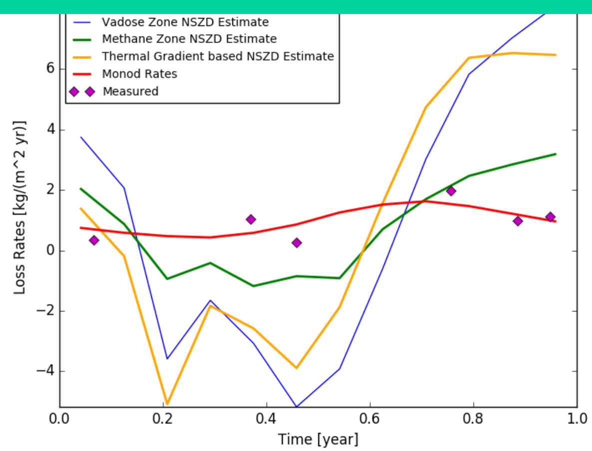




Short term Average Thermal Gradient NSZD rates



Monthly Average Thermal Gradient NSZD rates



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Model Output

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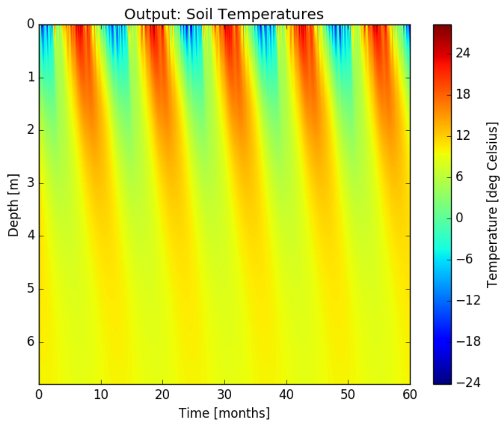
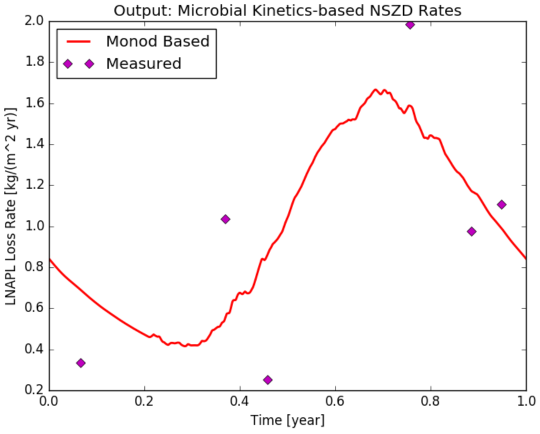
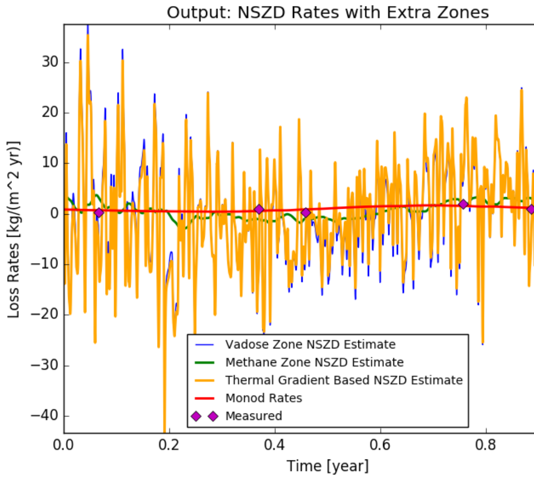
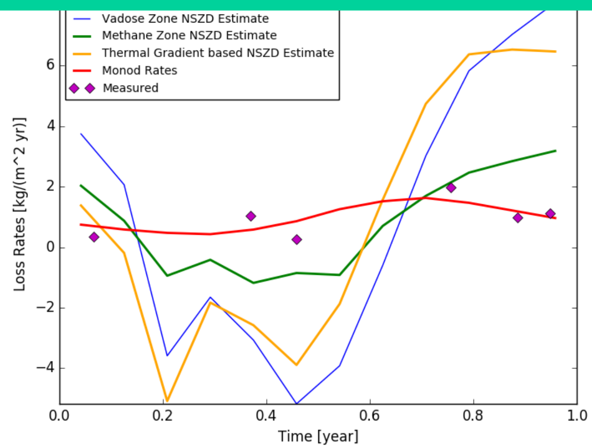






Monthly Average Thermal Gradient NSZD rates



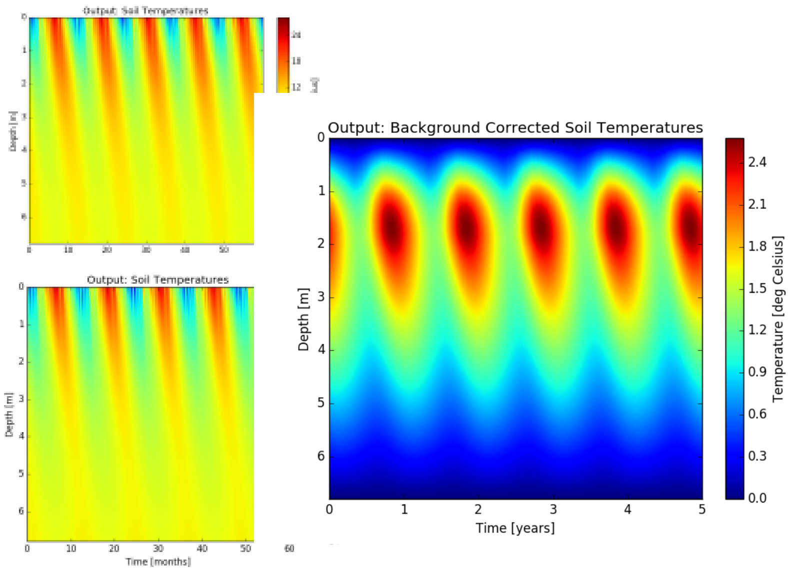
Annual Average Thermal Gradient NSZD rates

1. Thermal gradient location	Error Rate
Methane oxidation zone	**19%**
Aerobic Zone	**0.4%**
Entire Vadose Zone	**0.1%**

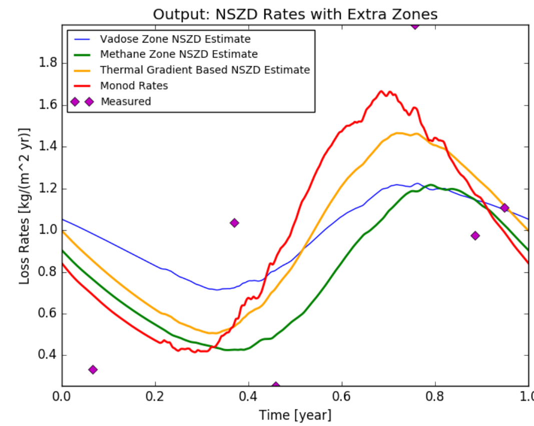
2 Ideal Background Correction

$\alpha_{site} = \alpha_{background} = 3.58 \times 10^{-07}\ m^2/s$

Model Output



Short term Average Thermal Gradient NSZD rates



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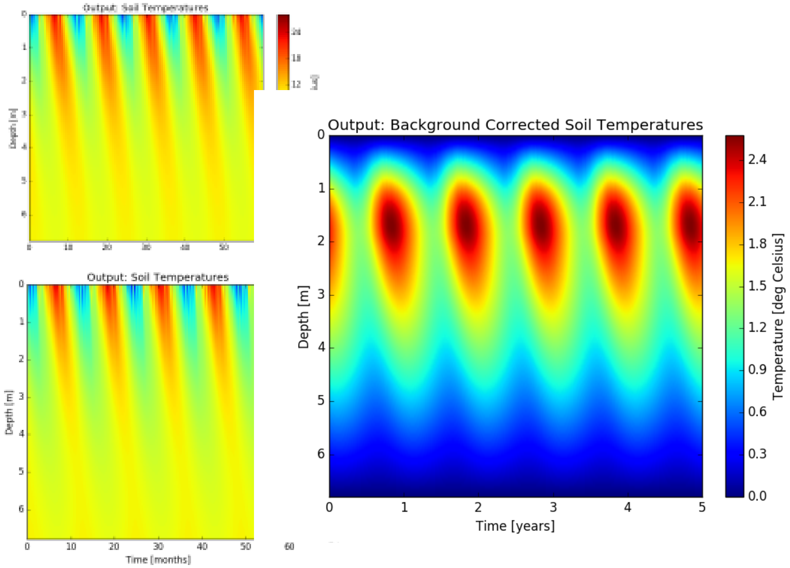

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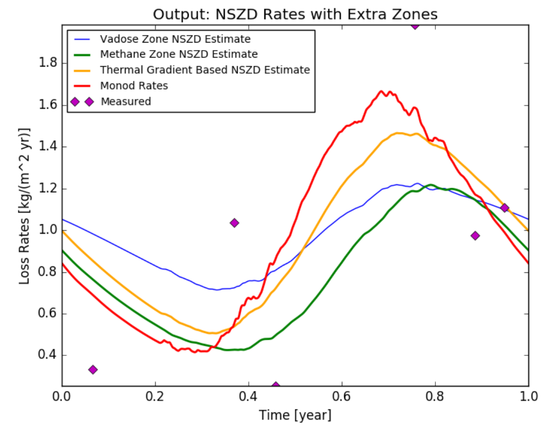

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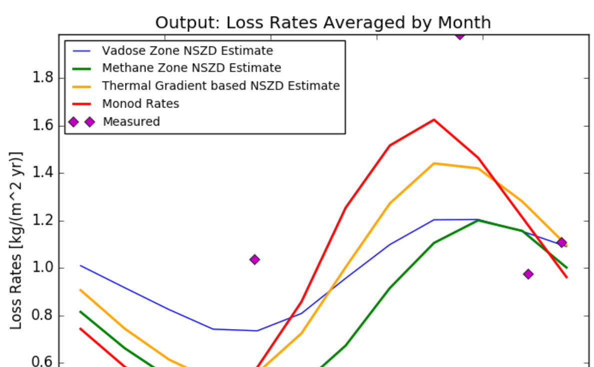
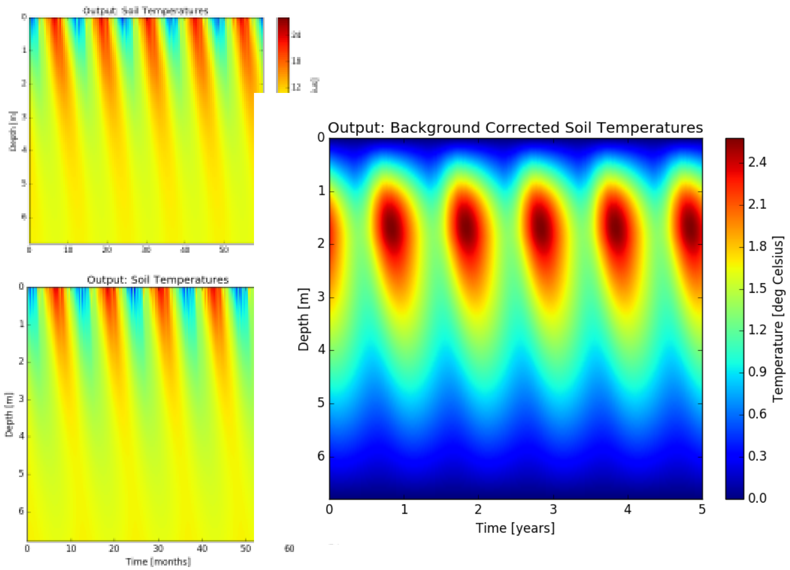


Ideal Background Correction

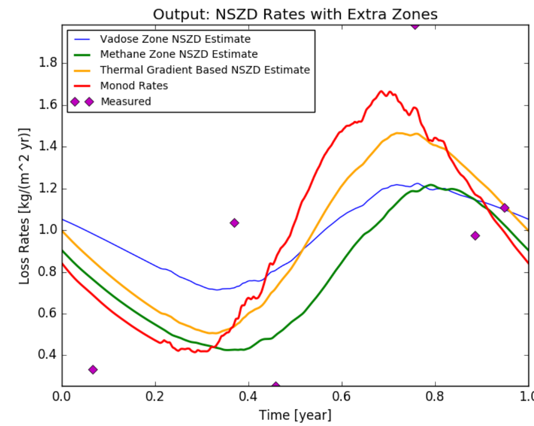
2

$\alpha_{site} = \alpha_{background} = 3.58 \times 10^{-07}\ m^2/s$

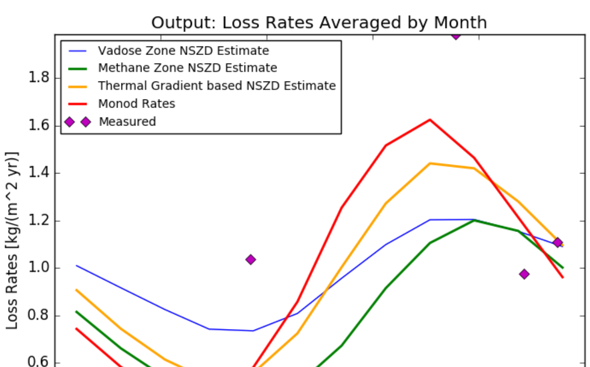
Model Output



Short term Average Thermal Gradient NSZD rates



Monthly Average Thermal Gradient NSZD rates



Annual Average Thermal Gradient NSZD rates

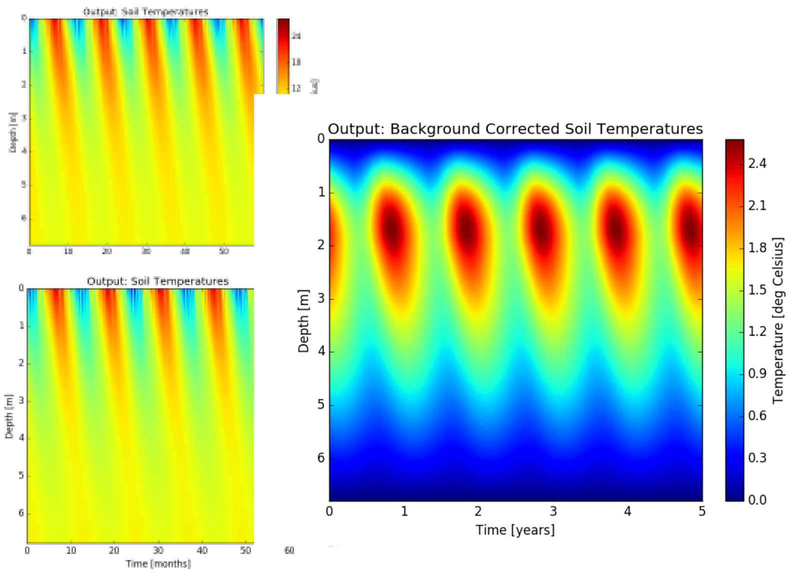
1. Thermal gradient location	Error Rate
Methane oxidation zone	**26.78%**
Aerobic Zone	**0.64%**
Entire Vadose Zone	**-0.57%**

Non-ideal Background Correction

3

α_{site} = 3.58 x 10 $^{-07}$ m^2/s , $\alpha_{background}$ =2 α_{site}

Model Output



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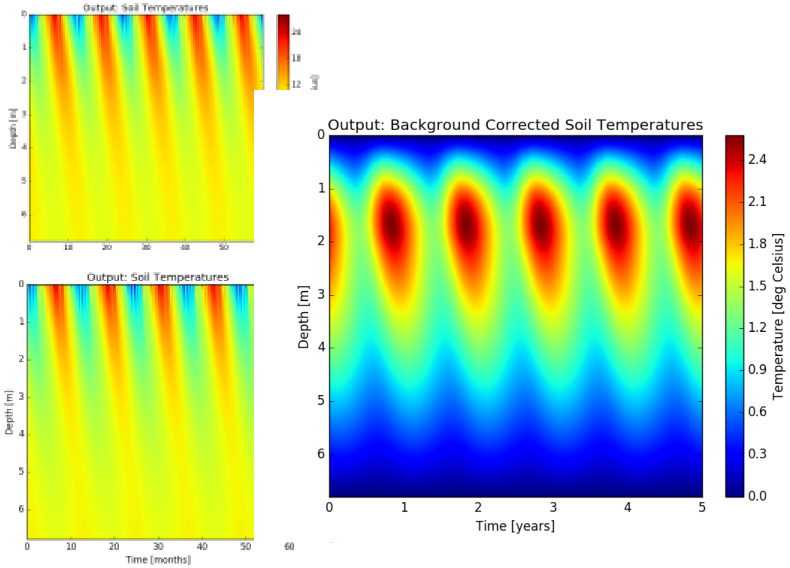
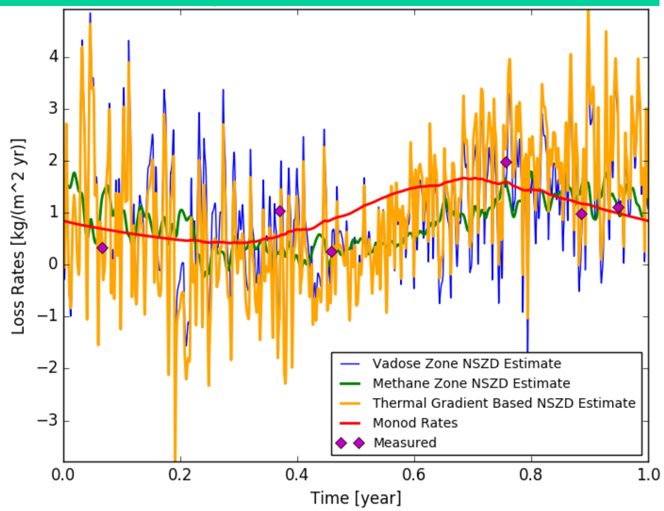


Short term Average Thermal Gradient NSZD rates



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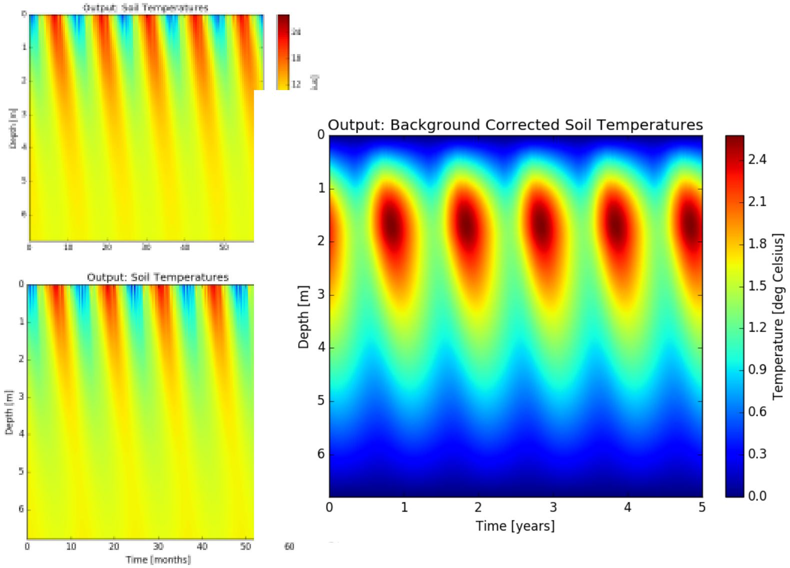

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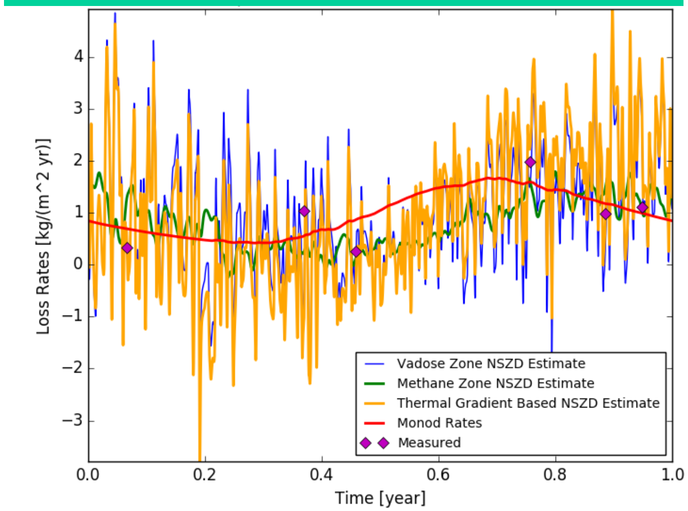

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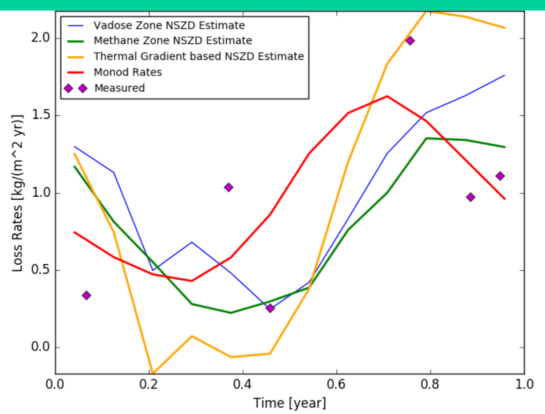

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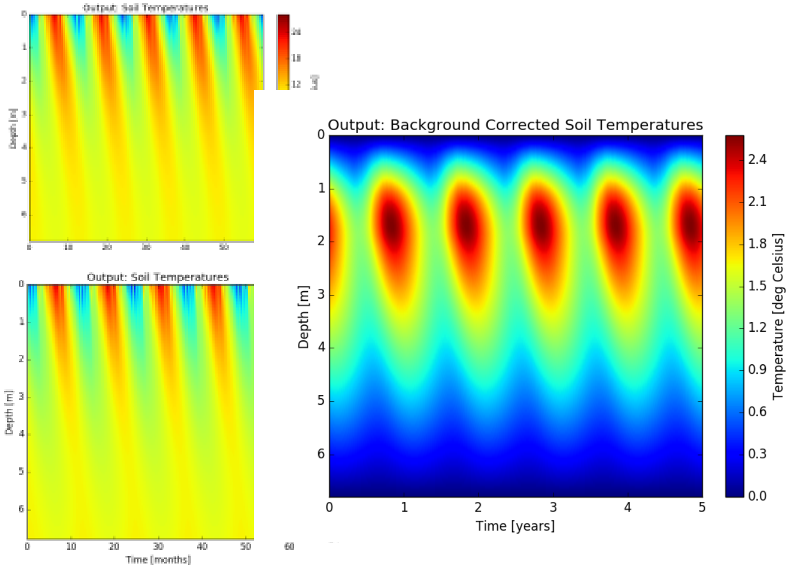

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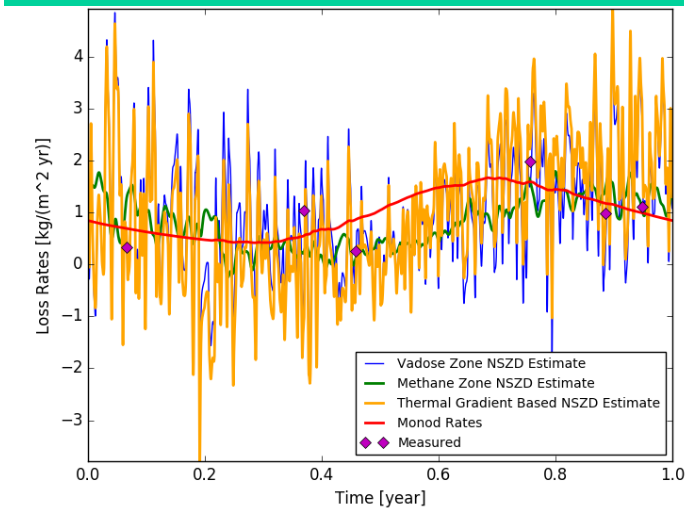
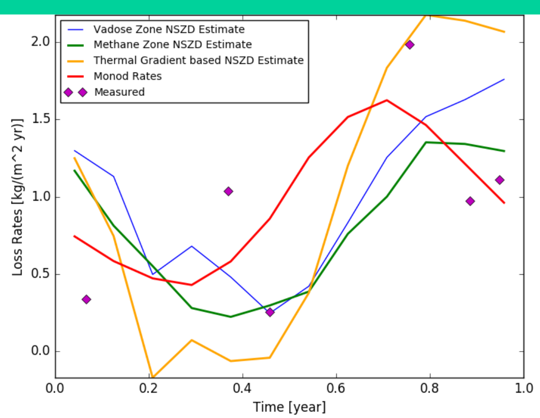


Monthly Average Thermal Gradient NSZD rates



Annual Average Thermal Gradient NSZD rates

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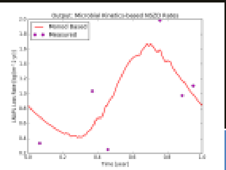
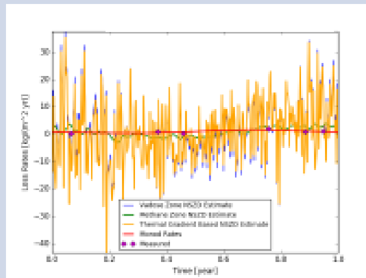
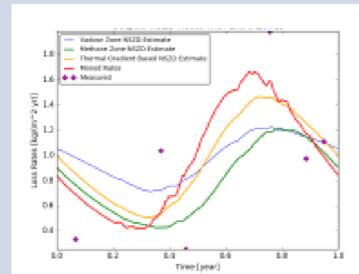
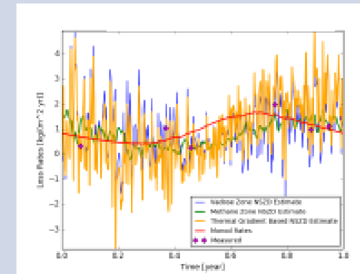
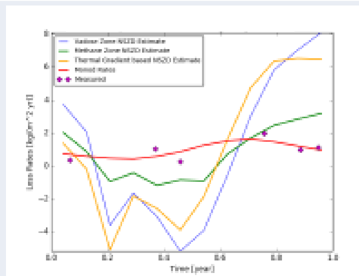
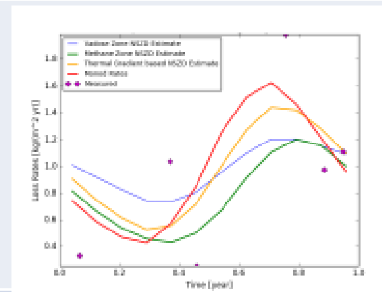
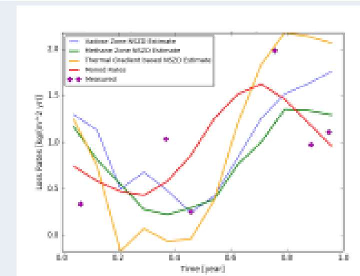
Entire Vadose Zone	**-0.57%**



Average Annual Thermal Gradients

	① Absolute temperatures	② Perfect Background	③ Imperfect Background
Short term			
Monthly Averages			
Annual Averages	Target: $R_{Monod,annual}$ = 0.97 kg/m2.yr = 1,200 gallons/ac.yr		
Methane Oxidation Zone	0.79 kg/m2.yr (19%)	0.788 (19%)	0.78 (19%)
Entire vadose zone	0.97 (0.4%)	0.97 (0.4%)	0.978 (0.4%)
Aerobic zone	0.96 (1%)	0.97 (1%)	0.96 (1%)

Further Reading on Long-Term Thermal

Thermal gradient method very sensitive to background location selection (Rayner et al, 2020)

Single Stick Method (Askarami and Sale, 2020) no background location if heat balances is cumulative (integrated through time)

Further Reading on Long-Term Thermal

Water Research 169 (2020) 115245

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Thermal estimation of natural source zone depletion rates without background correction

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A R T I C L E I N F O

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A B S T R A C T

Real-time monitoring of subsurface temperature profiles is a promising approach to resolving natural source zone depletion (NSZD) rates for shallow petroleum liquids. Herein, a new "single stick" computational method for transforming temperature data into NSZD rates is advanced. The method is predicated on subsurface temperatures being a function of surface heating and cooling, and the heat associated with NSZD. Given subsurface temperature at two points, a system of two-equation two-unknown is used to resolve NSZD rates. Mathematical formulations and computational algorithms are validated through computational tests showing near perfect agreement between prescribed and pre-

Askarami and Sale, 2020, Single Stick Method: Analytical Solution to the Heat Equation solved at each time step (i.e., daily), then numerically integrated through time.

Further Reading on Long-Term Thermal

- Battelle 2018 Conference

US 20170023539A1

(19) **United States**
(12) **Patent Application Publication**
Zimbron

(10) **Pub. No.: US 2017/0023539 A1**
(43) **Pub. Date: Jan. 26, 2017**

(54) ESTABLISHMENT OF CONTAMINANT DEGRADATION RATES IS SOILS USING TEMPERATURE GRADIENTS, ASSOCIATED METHODS, SYSTEMS AND DEVICES

(52) U.S. Cl.
CPC *G01N 33/24* (2013.01); *G01N 25/4846* (2013.01); *C12Q 1/04* (2013.01); *G01N 2033/243* (2013.01)

- Askarami and Sale, 2020

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Kayvan Karimi Askarani, Thomas Clay Sale*

Civil and Environmental Engineering Department, Colorado State University, 1320 Campus Delivery, B01, Fort Collins, CO. 80523-1320, USA

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ABSTRACT

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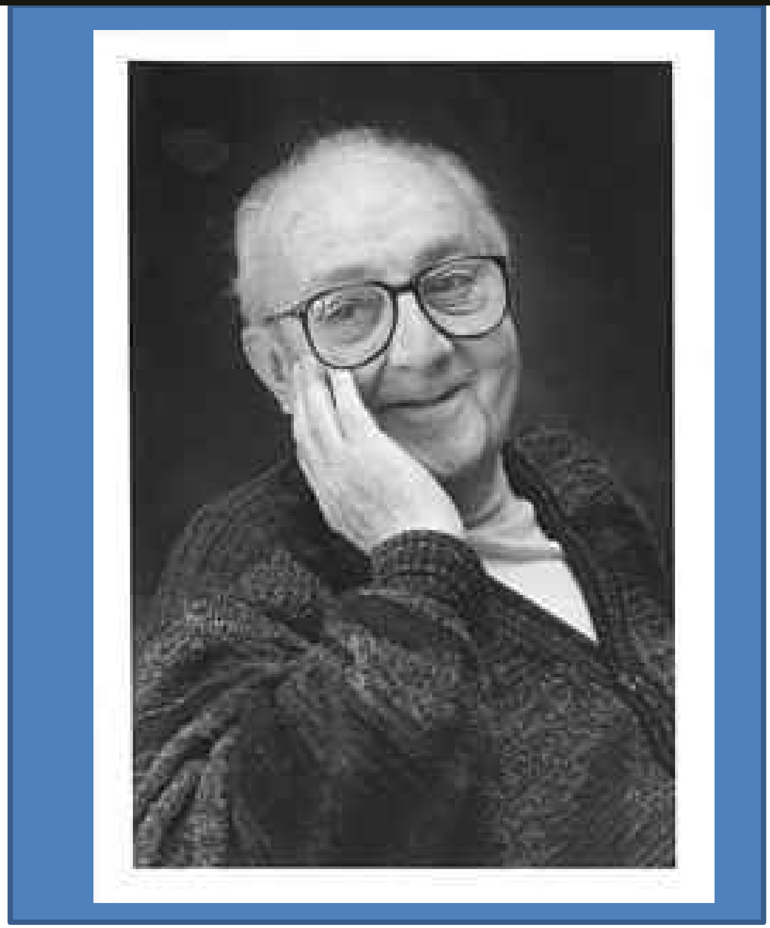
Thermal gradient method very sensitive to background location selection (Rayner et al, 2020)
Both long term approaches reduce to similar practice: long term heat balances reduces error

Conclusions

- Simple model improves understanding of LNAPL NSZD CSM
- Simulated temperature measurement errors do not seem large with respect to error due to short term ambient temperature fluctuations
- Ideal background location reduces error rates (short term and monthly averages)
- Departures from ideal background correction introduce significant errors
- Noise due to short-term fluctuations in ambient temperatures cancels out over an annual (seasonal cycle) period
- Annual averaging improves thermal gradient-based LNAPL loss rate estimates within 1% or less target, as long as locations chosen are outside reactive zone: (*US Pat. 62/151.564*)

On models…



(George E. P. Box, 1987)

"Essentially, all models are wrong, but some are useful"

"…the practical question is how wrong do they have to be to not be useful"

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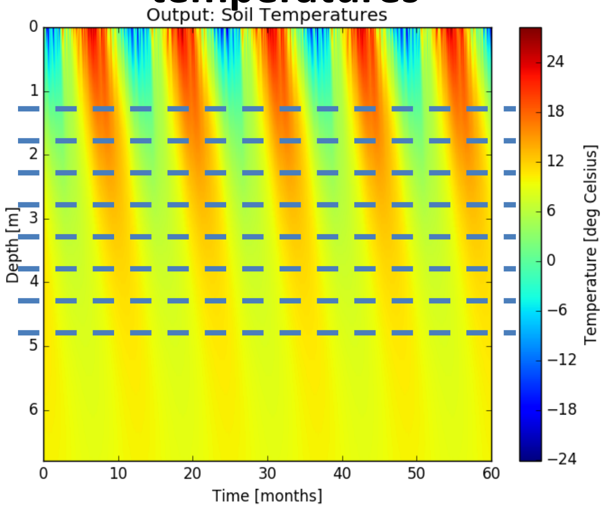
Thermal Gradient Effects of $T_{\text{Measurement Error}}$

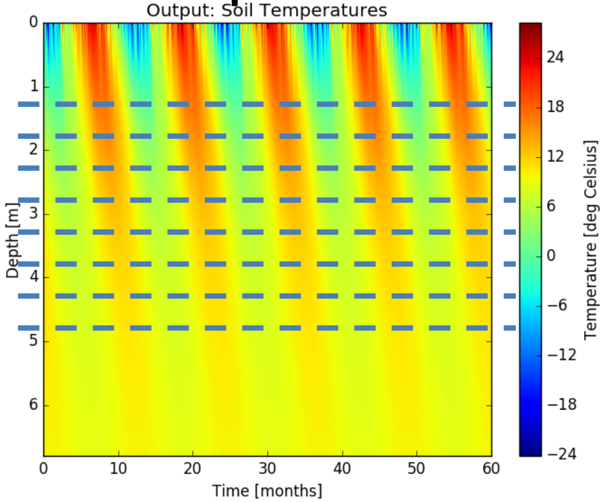
Ideal soil temperatures



i) Sensor location ±(2.5cm)

Thermal Gradient Effects of T Measurement Error

Ideal soil temperatures

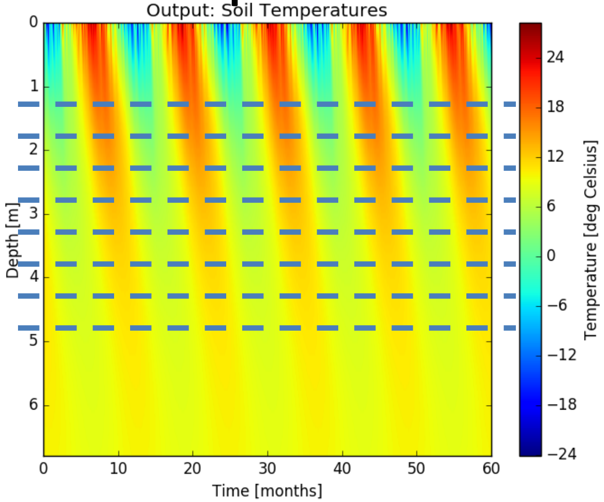


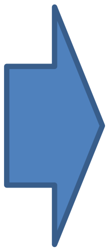
i) Sensor location

ii) Simulate thermocoluple spacing (16 TCs on 8 m, 0.5 m appart)

Thermal Gradient Effects of $T_{\text{Measurement Error}}$

Ideal soil temperatures





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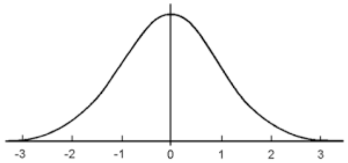

iii) Add T measurement error (per assumed manufacturer spec)

Thermal Gradient Effects of $T_{\text{Measurement Error}}$

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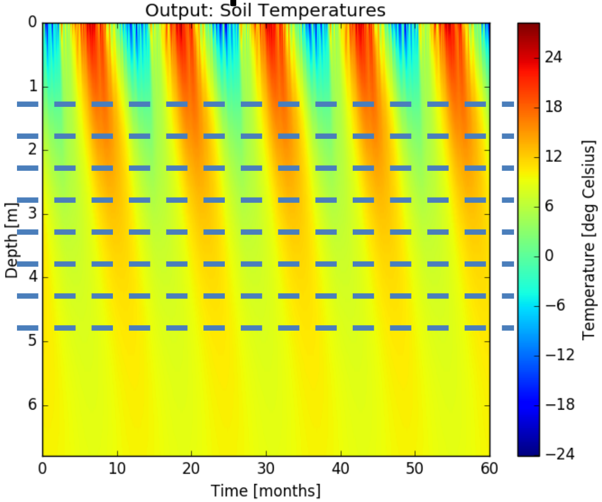

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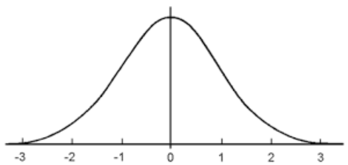

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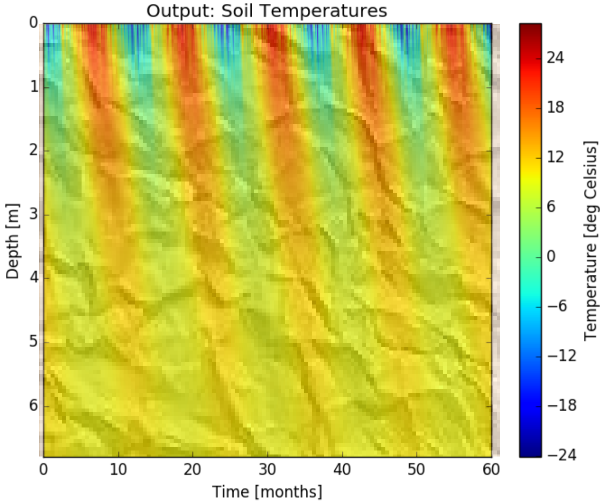
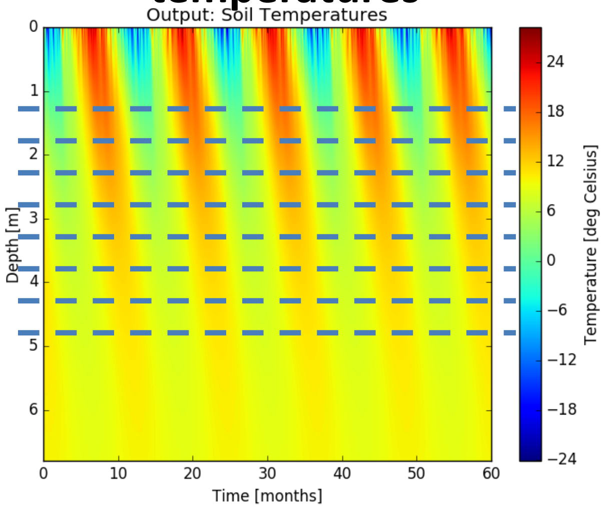
Simulated measured soil temperatures



Thermal Gradient Effects of T Measurement Error

Ideal soil temperatures



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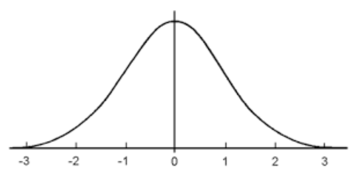

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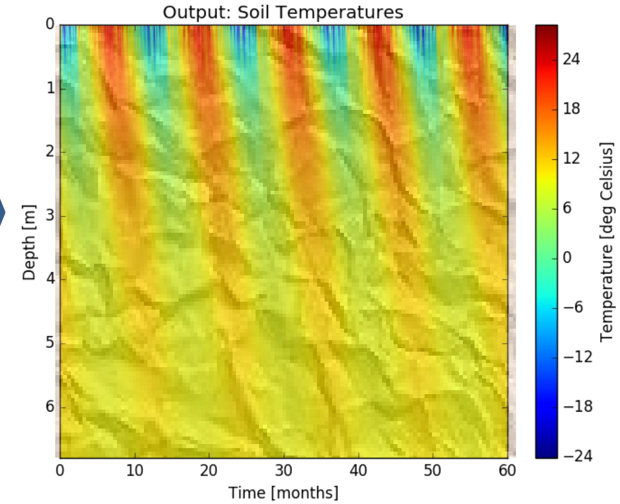
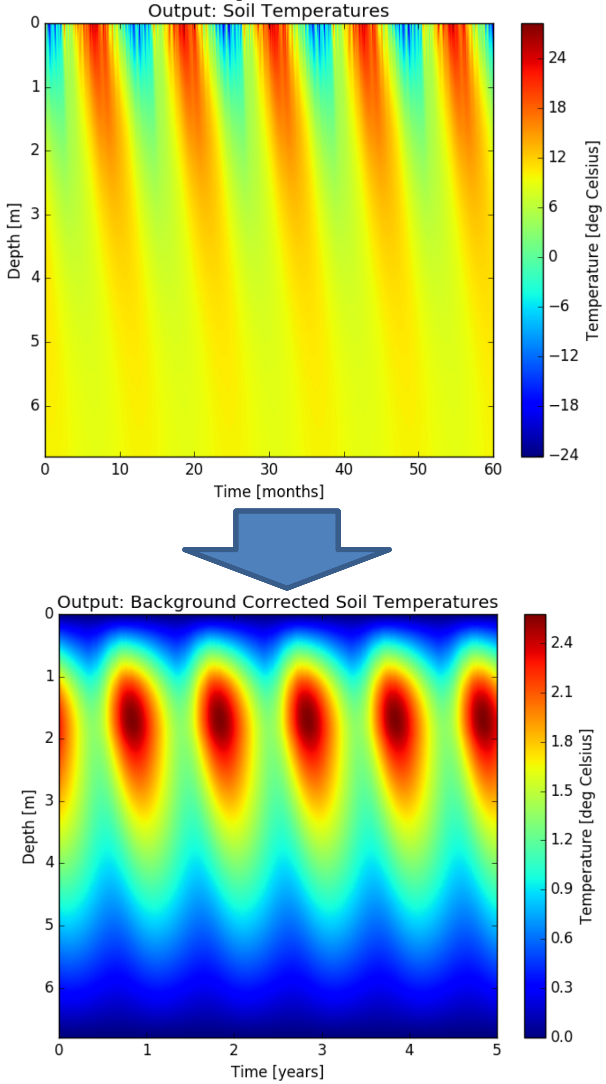


Thermal Gradient Method

Thermal Gradient Effects of T Measurement Error

Ideal soil temperatures



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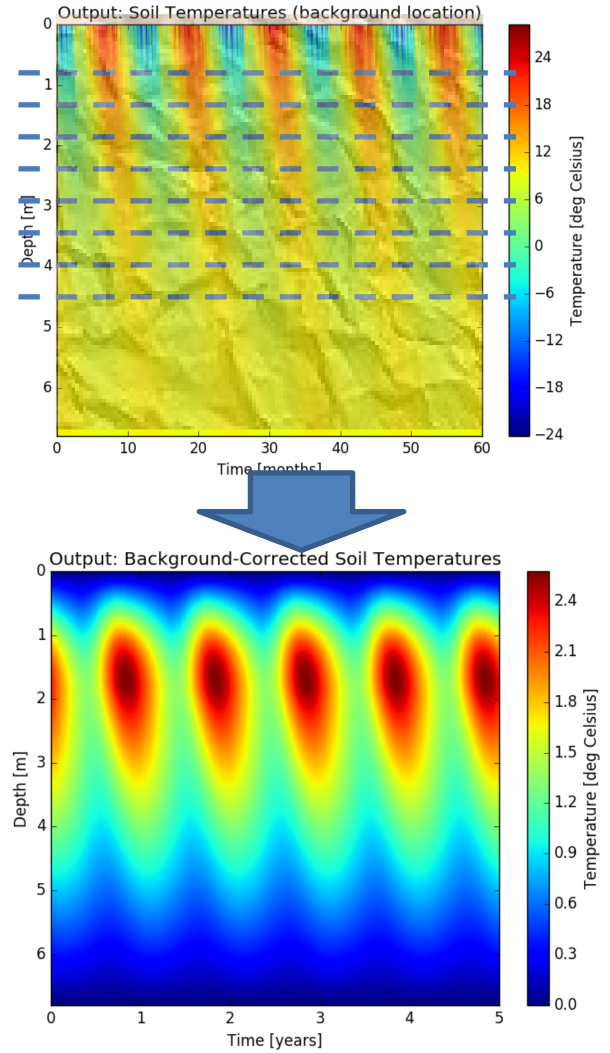
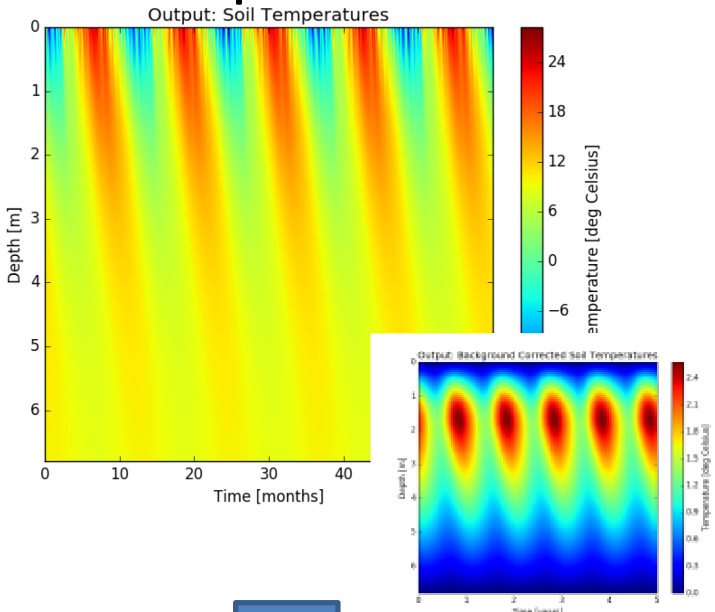
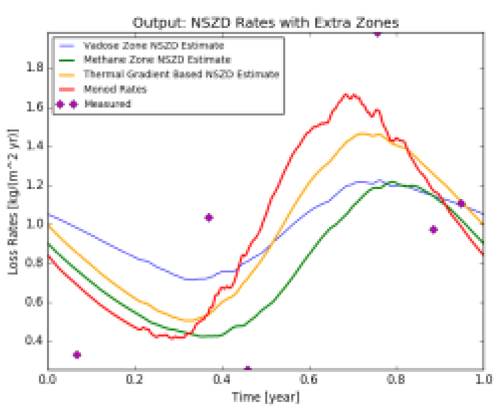


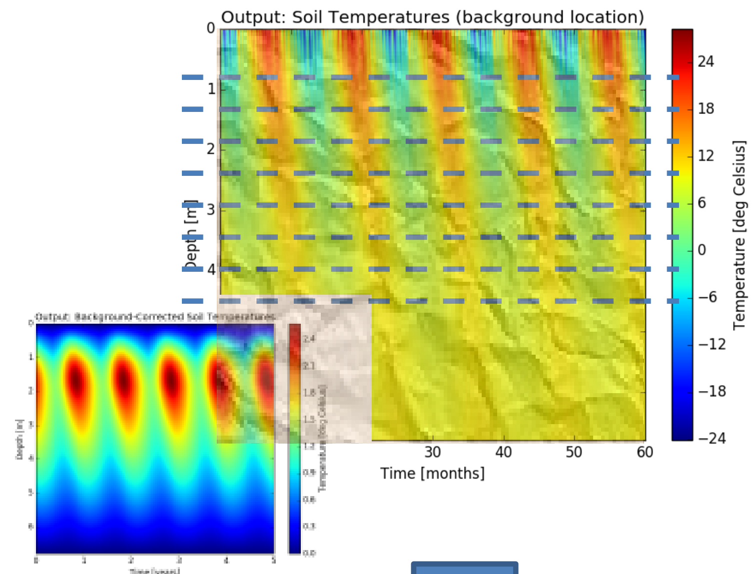
Thermal Gradient Effects of T Measurement Error

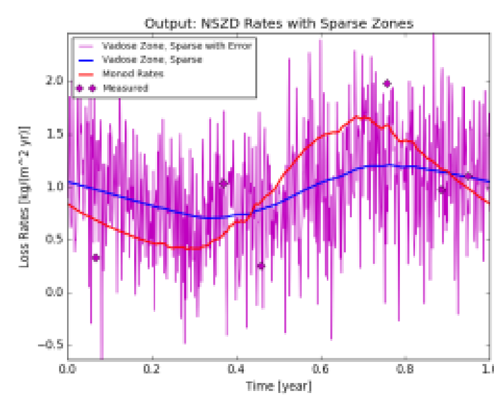
Ideal soil temperatures





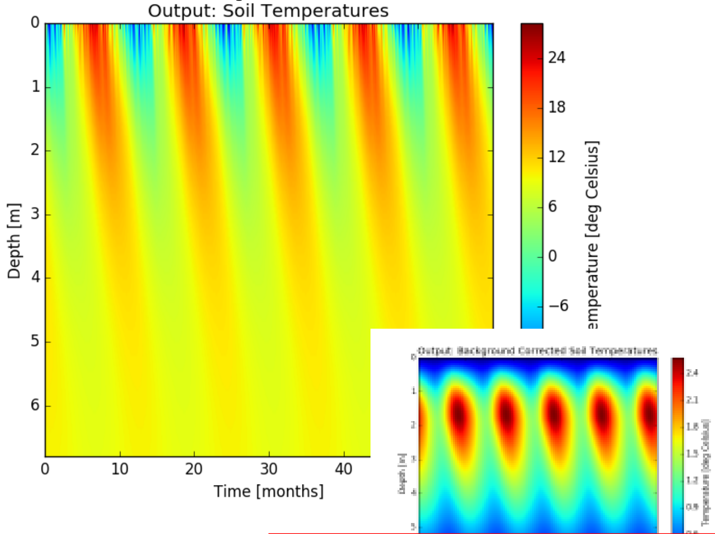
Simulated measured soil temperatures



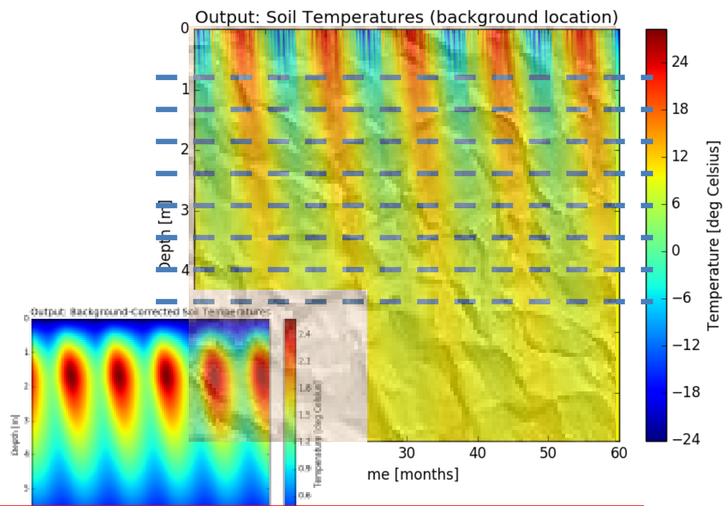


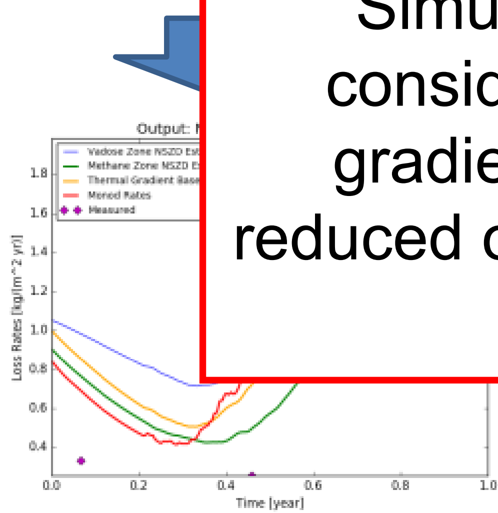
Thermal Gradient Effects of T Measurement Error

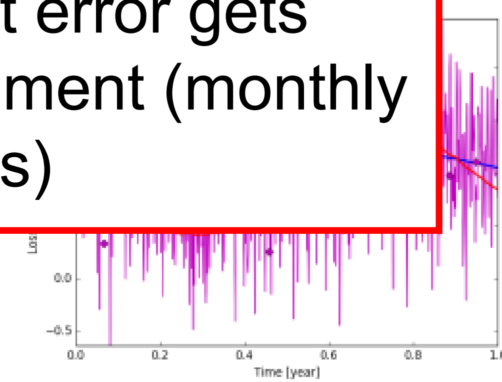
Ideal soil temperatures



Simulated measured soil temperatures





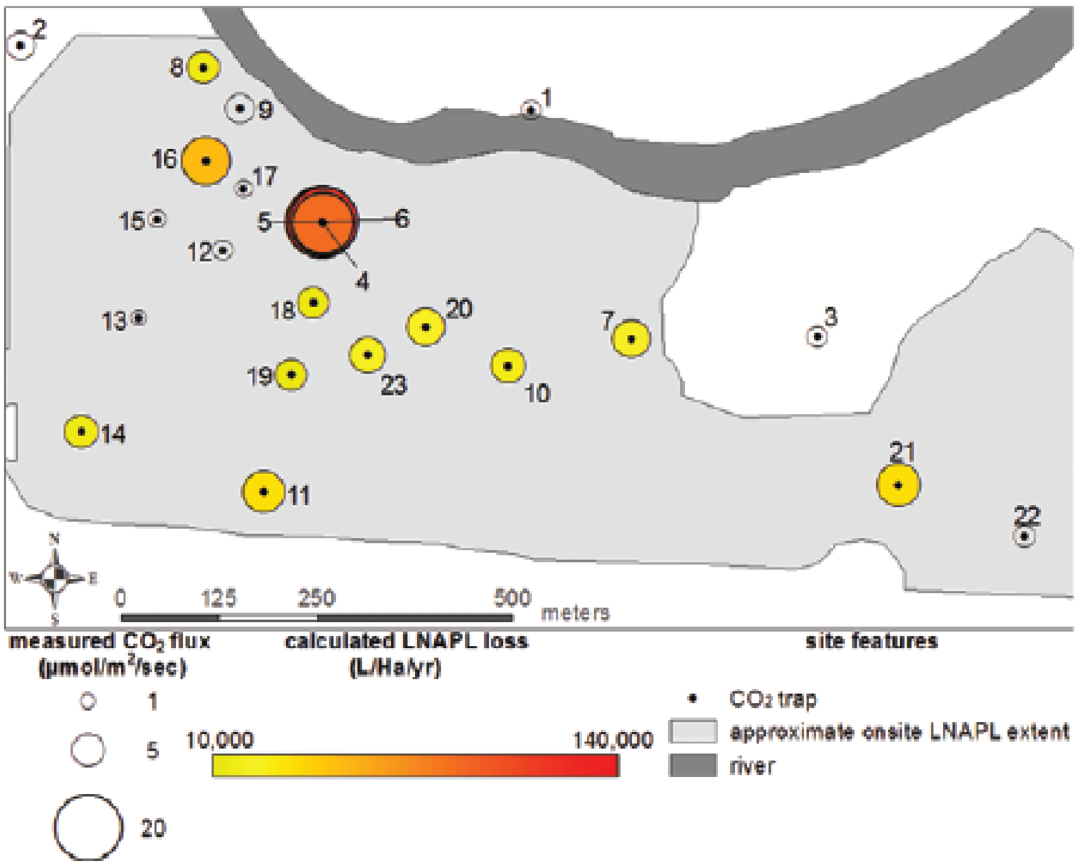


Simulated noisy soil temperatures add considerable error to short term thermal-gradient NSZD estimates, but error gets reduced over long term measurement (monthly and annual averages)

Questions?

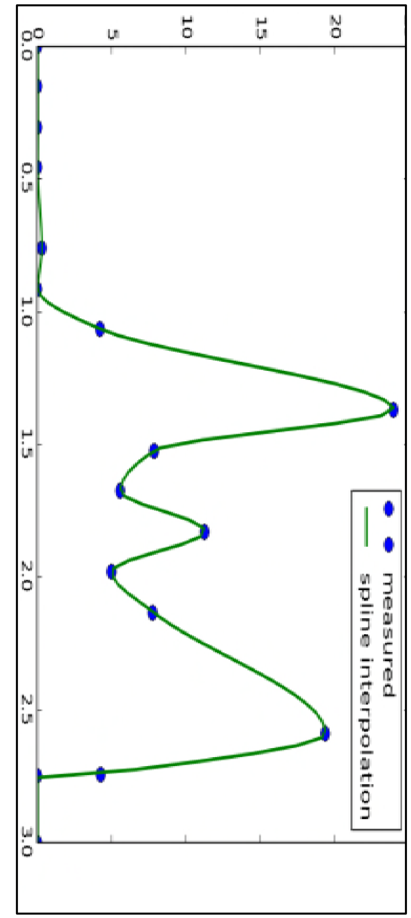
Case Study 2: Former refinery

- Depth to Groundwater: 3 m
- Average Groundwater Temperature: 14 °C



Source: McCoy et al., 2014

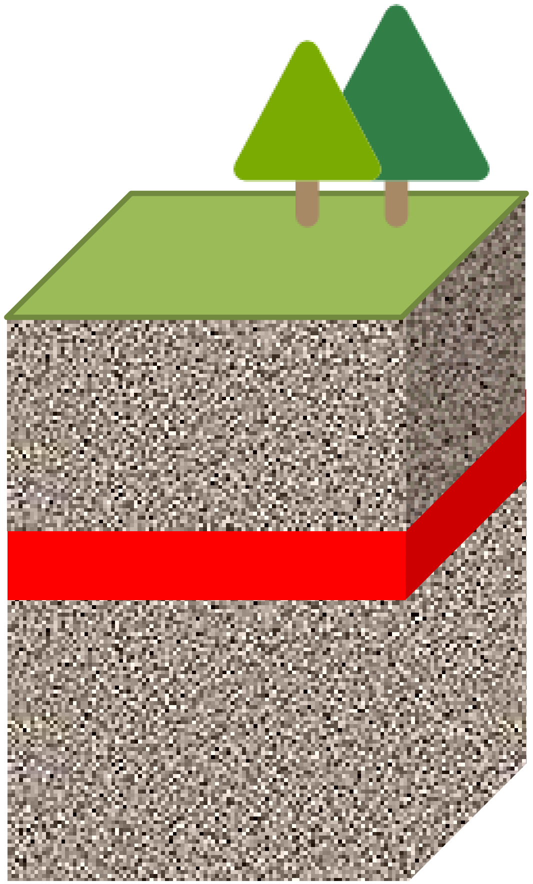
LNAPL concentration (kg/m^3)



Irianni-Renno, 2014

Methods for Estimating LNAPL Loss

Using thermal gradients



Methods for Estimating LNAPL Loss

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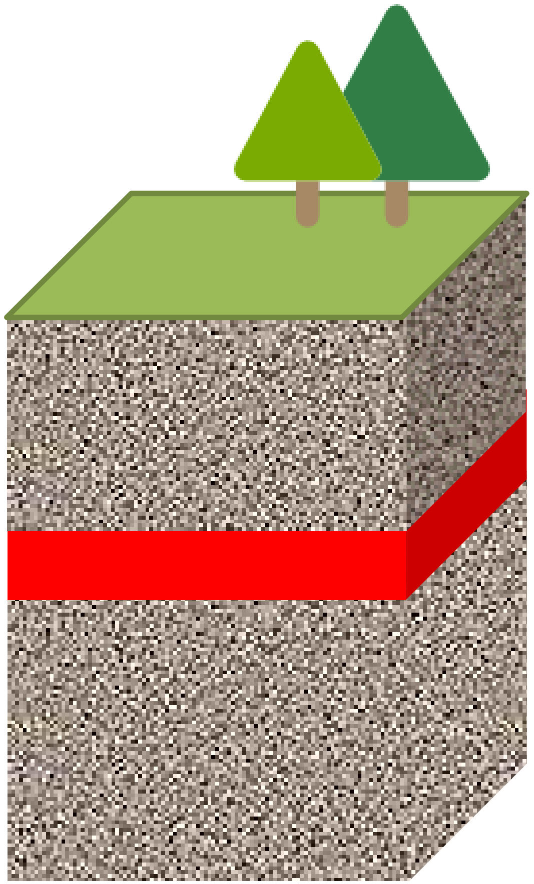
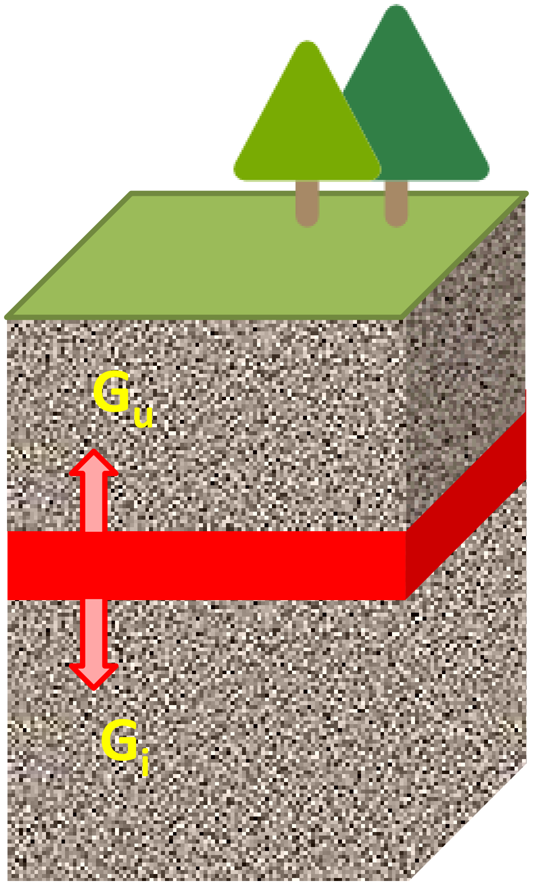


Methods for Estimating LNAPL Loss

Using thermal gradients

$$G = G_u + G_i$$



Methods for Estimating LNAPL Loss

Using thermal gradients

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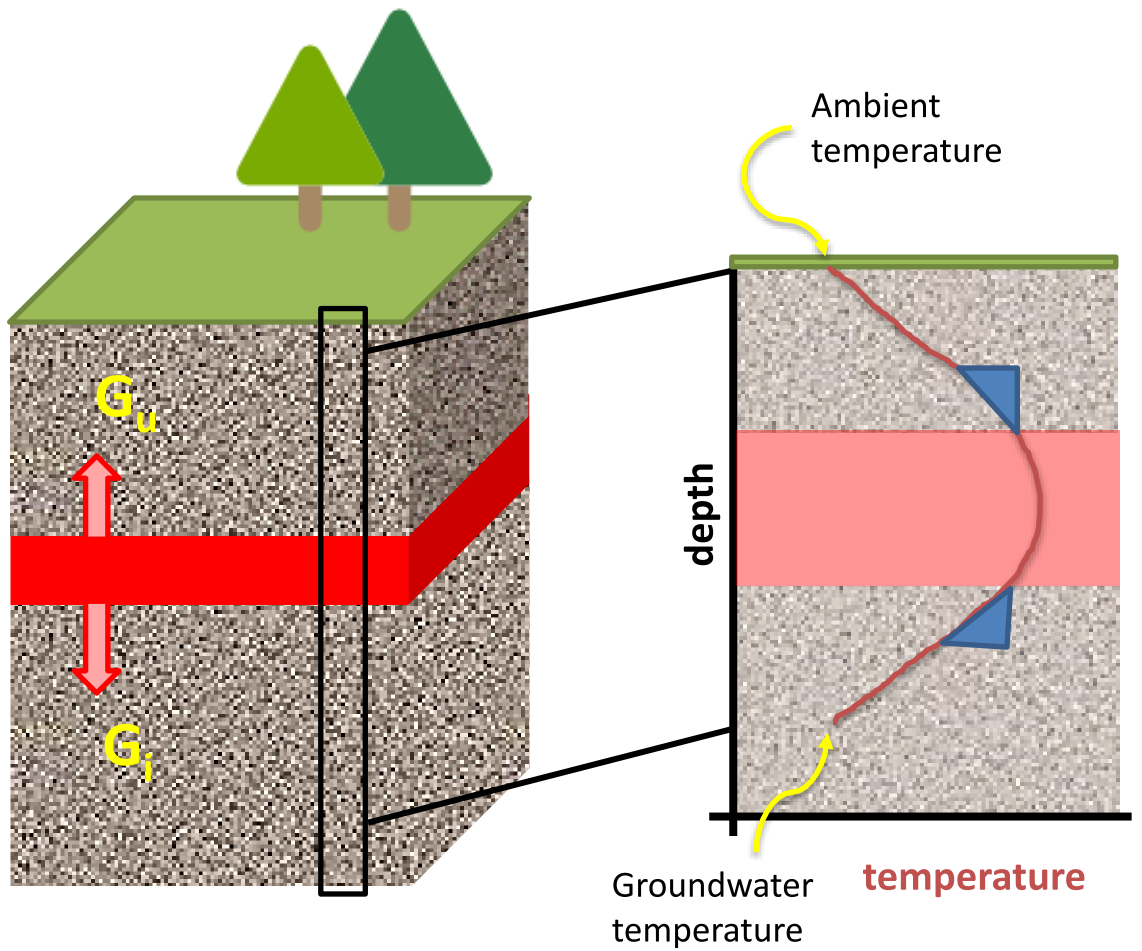

Modeling Heat Transfer in Soils

Laws of Nature

Modeling Heat Transfer in Soils

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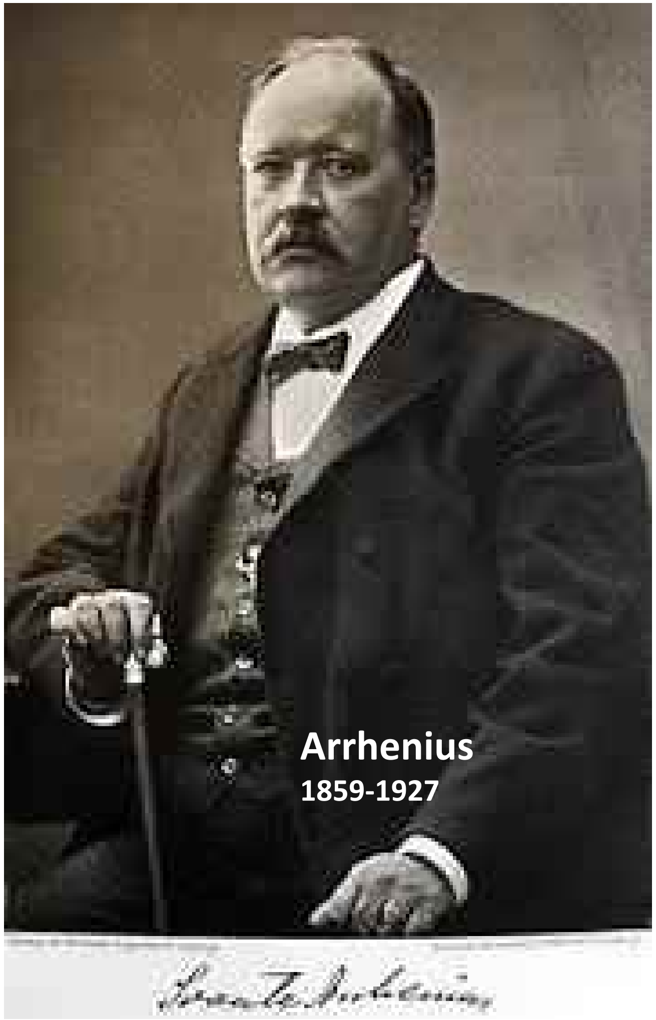
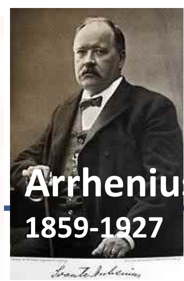


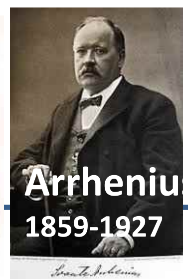
Modeling Heat Transfer in Soils

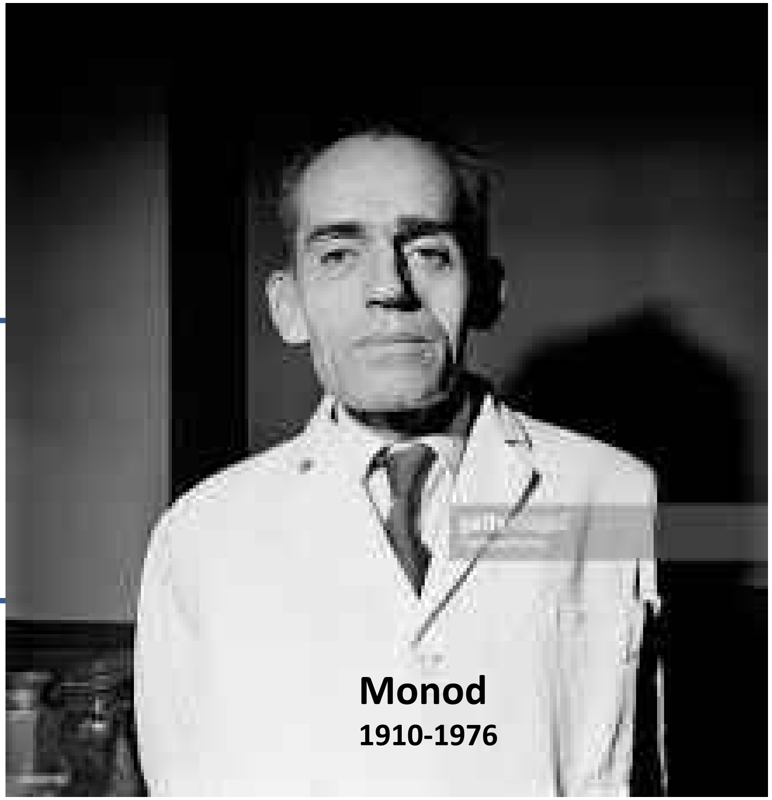
Laws of Nature



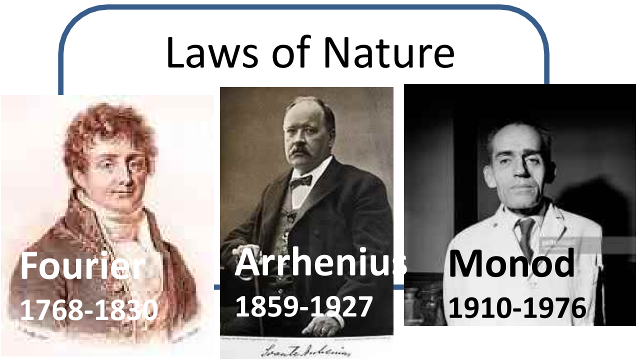
Modeling Heat Transfer in Soils

Laws of Nature





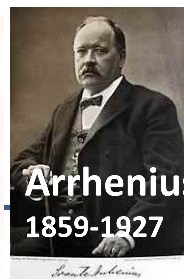
Modeling Heat Transfer in Soils

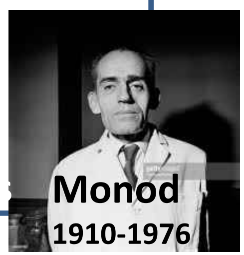


Modeling Heat Transfer in Soils

Field Data

Laws of Nature





Modeling Heat Transfer in Soils

Field Data

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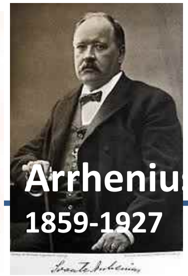

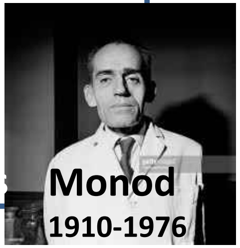

Modeling Heat Transfer in Soils

Field Data

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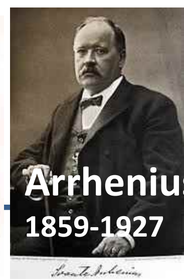
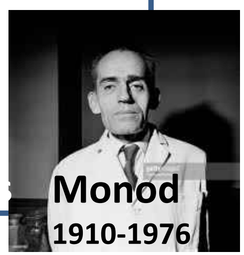


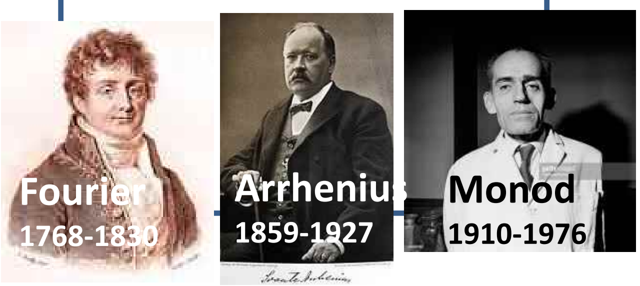


A simple, yet realistic geometry

Modeling Heat Transfer in Soils

Field Data

Laws of Nature



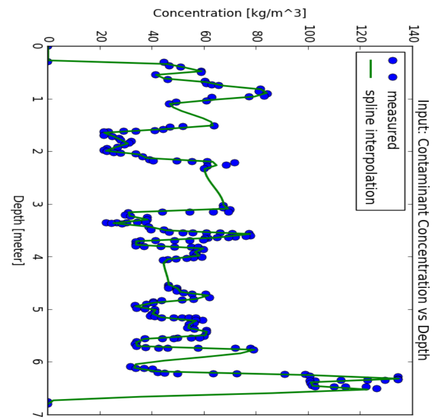
Bio∘Therm

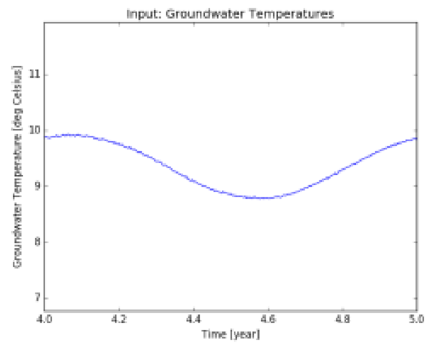
A simple, yet realistic geometry

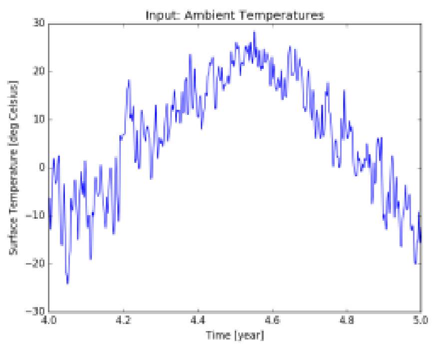
Available at:

www.BiogenicHeat.com

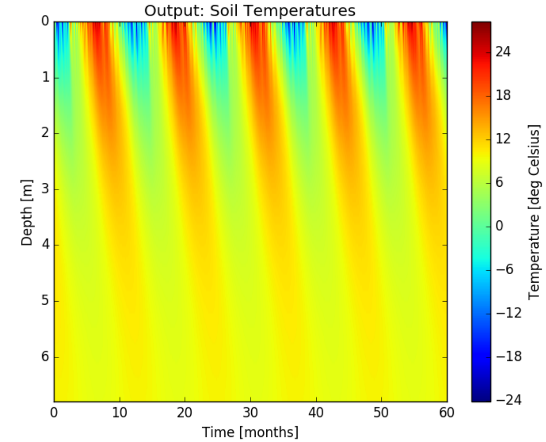
Inputs and Outputs

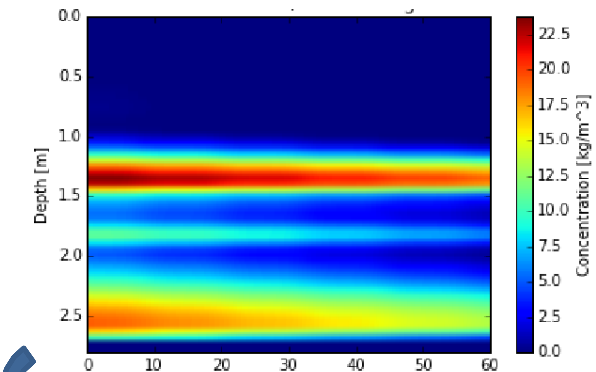


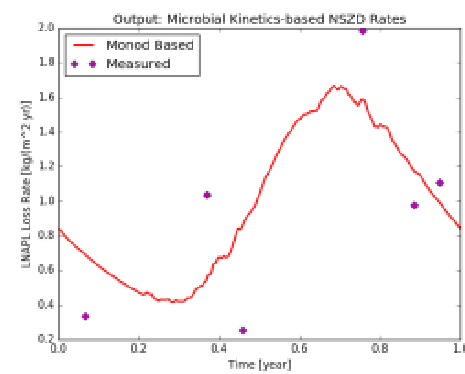




BioTherm

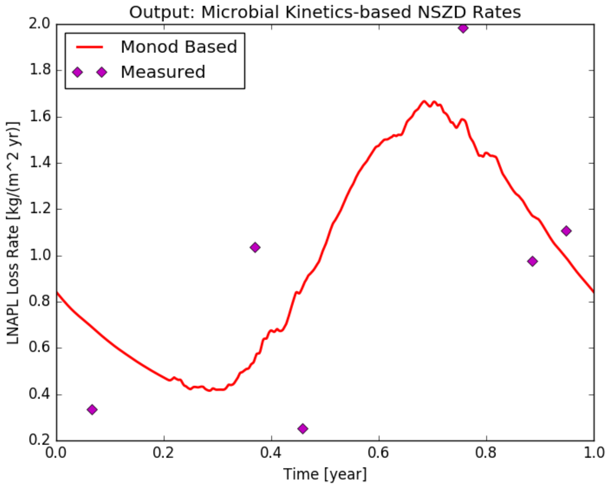






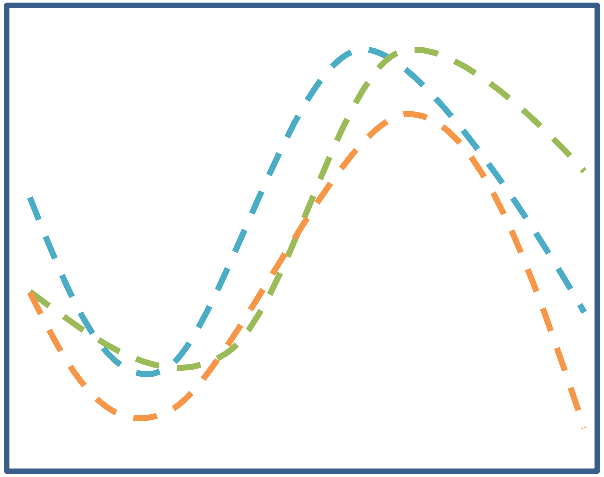
Can both approaches be reconciled by Bio∘Therm ?

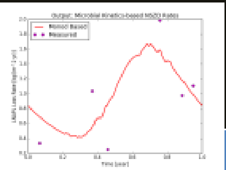
1. Using the mass balance/ Monod rates



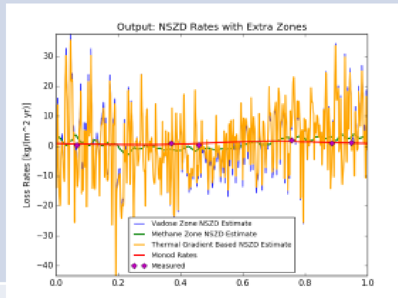
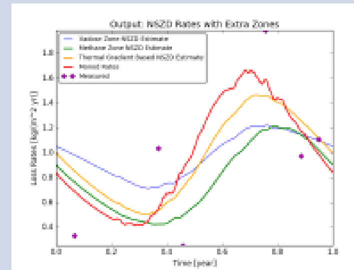
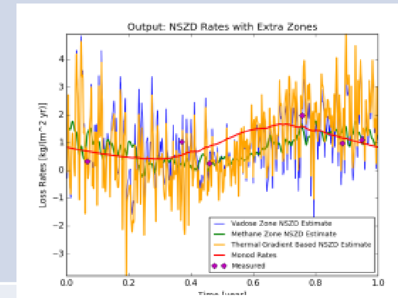
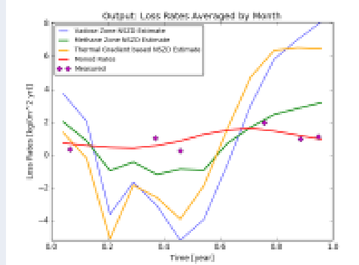
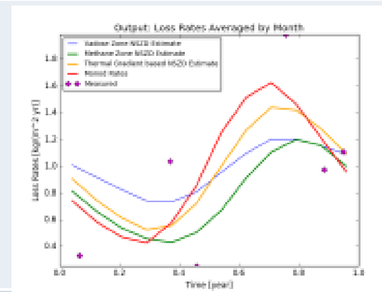
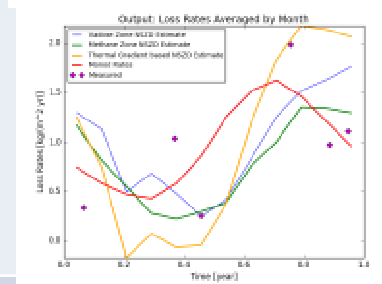
Vs.

2. Using thermal gradients





Average Annual Thermal Gradients

	1 Absolute temperatures	2 Perfect Background	3 Imperfect Background
Short term			
Monthly Averages			
Annual Averages	Target: $R_{Monod,annual}$ = 0.97 kg/m2.yr = 1,200 gallons/ac.yr		
Methane Oxidation Zone	0.79 kg/m2.yr (19%)	0.788 (19%)	0.78 (19%)
Entire vadose zone	0.97 (0.4%)	0.97 (0.4%)	0.978 (0.4%)
Aerobic zone	0.96 (1%)	0.97 (1%)	0.96 (1%)