Mathematical Modelling of Atmospheric Contaminant Dispersion

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Acknowledgments

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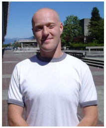
- Funded jointly by MITACS and Teck-Cominco



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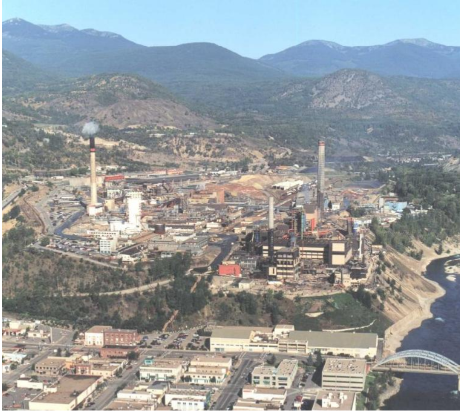
- Sup't, Environmental Management

Motivation

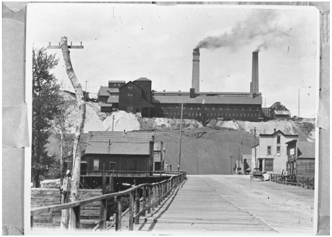
- Teck-Cominco operates one of the world's largest integrated lead-zinc smelting operations in Trail BC on the Columbia River.

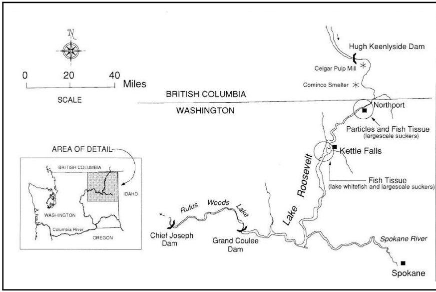
- It's a major driver of BC economy but also one of Canada's biggest polluters (lead, zinc, arsenic, cadmium, mercury, SO_2, ...).
- Annual emissions reporting is required under the *National Pollutant Release Inventory* (`http://www.ec.gc.ca`).



Historical Sidebar

- Trail was founded in 1890s as a mine supply point. A small smelter was built.
- In 1906, Cominco was formed.
- In 1941, WA state was awarded damages from Cominco for trans-border pollution – one of the most-cited international law cases.
- From 1917 to 1940, emissions of SO_2 were 100–700 T/day. Currently down to 22 T/yr.
- Today Teck-Cominco prides itself on its "clean" Trail operations.





The Problem

- Teck's Trail operation is unable to directly measure stack emissions in a reliable or cost-effective manner.
- Teck-Cominco's reporting has so far relied on "engineering estimates" based on various chemical processes.
- Measurements of the following particulates have been taken using "dustfall jars" during 2001–2002:

  zinc (Zn), sulphates (x-SO_4) and strontium (Sr).

Key Questions:

Is there a robust method that will provide *reliable* estimates of stack emission rates based on deposition measurements?
And what is "reliable"? . . . errors of 25-50% in estimates are considered acceptable, as long as they are overestimates!

Outline

Atmospheric Dispersion

- "Atmospheric disperion" refers to transport of contaminants via two processes:
  1. advection by the wind, and
  2. turbulent diffusion.
- Reduces to solving the advection-diffusion equation

$$\frac{\partial C}{\partial t} + \vec{u} \cdot \nabla C = \nabla \cdot (K \nabla C) + Q - R$$

where

$$\begin{aligned} C(\vec{x}, t) &= \text{concentration (or density) of the contaminant } (kg/m^3) \\ \vec{u}(\vec{x}, t) &= \text{given wind velocity } (m/s) \\ K &= \text{turbulent eddy diffusivity } (m^2/s) \\ Q &= \text{emission source term } (kg/m^3\, s) \\ R &= \text{sink term from reactions, etc. } (kg/m^3\, s) \end{aligned}$$

Atmospheric Dispersion 2

- A variety of approaches have been used for the solving the advection-diffusion equation, including
  - *analytical*: asymptotics, Green's functions.
  - *computational*: finite difference, finite volume, spectral.
- Scales of interest range from

  $$10 \text{ m} \rightarrow 100 \text{ km} \qquad \text{and} \qquad \text{minutes} \rightarrow \text{months},$$

  ... making many methods impractical, particularly in 3D.
- Most industry-standard software is based on Gaussian plume solutions (ref: `epa.gov`, "recommended models").
- Most work has focused on solving the forward problem:

> *Given a set of source emission rates, calculate deposition*

as opposed to the inverse problem:

> *Given a set of deposition values, calculate emission rates*

Simplifying Assumptions

Typical assumptions for a single, isolated stack:

- Stack is a point source located at $(0, 0, H)$.
- Constant emission rate Q $(kg/m^3\,s)$.
- Constant wind velocity U (m/s) in x-direction.
- Particles settle due to gravity at speed W_{set} (m/s).
- Deposition occurs at the ground at speed W_{dep} (m/s).
- Turbulent eddy diffusivities K_x, K_y, K_z (m^2/s) are constant.
  Turbulent eddy diffusivities K_x, K_y, K_z (m^2/s) are constant.
- Diffusion downwind is negligible compared to convection $(K_x = 0)$.
- Chemical reactions are negligible once particles are released.
- Variations in topography are ignored (rectangular domain).
- No inversions $(z \to \infty)$ and no confinement $(x, y \to \pm\infty)$.
- Steady state. Steady state – we'll relax this later.

Governing Equations

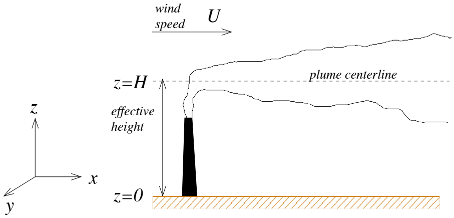
The advection-diffusion equation reduces to:

$$\underbrace{U\frac{\partial C}{\partial x}}_{\text{wind}} - \underbrace{W_{set}\frac{\partial C}{\partial z}}_{\text{settling}} = \underbrace{\frac{\partial}{\partial y}\left(K_y\frac{\partial C}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z\frac{\partial C}{\partial z}\right)}_{\text{cross-wind and vertical diffusion}} + \underbrace{Q\,\delta(x)\,\delta(y)\,\delta(z-H)}_{\text{point source}}$$

Boundary conditions:

$$K_z\frac{\partial C}{\partial z} + W_{set}C = W_{dep}C \qquad \text{at } z=0 \text{ (deposition)}$$

$$C \to 0 \qquad \text{as } x, y \to \pm\infty \text{ and } z \to \infty$$



Outline

Derivation

- Assume no settling or deposition ($W_{set} = W_{dep} = 0$)

$$\Longrightarrow \quad \text{Flux BC reduces to} \quad \frac{\partial C}{\partial z}(x,y,0) = 0$$

- Separable solution: $C(x,y,z) = A(x,y)\,B(x,z)$.
- Rescale variables: $X = \frac{1}{UH^2}\int_0^x K(x')\,dx'$, $Y = \frac{y}{H}$, $Z = \frac{z}{H}$.
- Yields two 2D diffusion problems:

$$\frac{\partial A}{\partial X} = \frac{\partial^2 A}{\partial Y^2}$$
$$A(0,Y) = C_o\delta(Y)$$
$$A(X,\pm\infty) = 0$$

$$\frac{\partial B}{\partial X} = \frac{\partial^2 B}{\partial Z^2}$$
$$B(0,Z) = C_o\delta(Z-1)$$
$$\frac{\partial B}{\partial Z}(X,0) = 0, \quad B(X,\infty) = 0$$

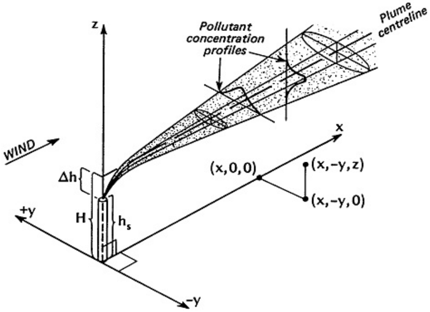
Gaussian Plume Solution

Use Laplace transform to obtain Gaussian plume (GP) solution:

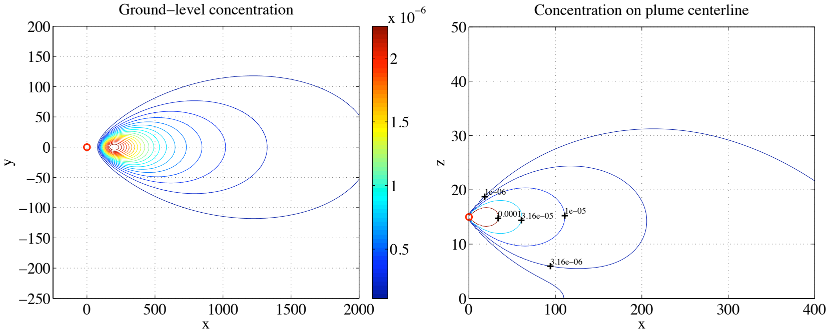
$$C(x,y,z) = \frac{Q}{2\pi U \sigma_y \sigma_z} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \left[\exp\left(-\frac{(z-H)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+H)^2}{2\sigma_z^2}\right)\right]$$

Note: Eddy diffusivities are replaced by standard deviations of concentration:

$$\sigma_{y,z}^2(x) = 2xK_{y,z}/U.$$



Typical Solution Contours



Notice peak in ground-level concentration downwind of source.

Practical Limitations

1. **Choice of σ:**
   - In practice, $\sigma = ax^b$ with a and b fit to observations (Brookhaven or Briggs formulas).
   - Only $b = \frac{1}{2}$ is consistent with GP solution since $K = \sigma^2 U/2x =$ constant.
   - Other exponents lead to errors in mass conservation, but these errors are only significant at long range (Winges, 1990).
2. **Errors at short and long range:**
   - GP solution is singular as $x \to 0$ (limits accuracy near source).
   - Errors grow at large x, but our domain is small (only 2 km^2).
3. **Calm winds:**
   - Common misconception: GP breaks down as $U \to 0$ **Wrong!**
   - In fact, $C \sim \frac{Q}{2\pi x\sqrt{K_y K_z}} + \mathcal{O}(U)$ – heat equation solution.
   - Real problem is an increase in plume rise (H) as $U \to 0$.
   - **Rule of thumb:** calm winds are defined as $U = 0.5$ m/s (Hanna et al., 1982).

Practical Limitations 2

4. **Steady assumption:**
   - GP is only strictly valid at steady state, but is a good estimate of average concentrations over long enough time intervals.

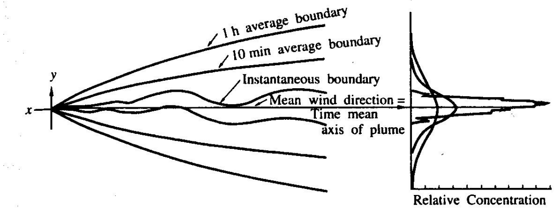


FIGURE 17.2 Plume boundaries and concentration distributions of a plume at different averaging times.

   - A 10 min. averaging time is consistent with observations (Hanna et al., 1982).
   - Also suggests that time-dependent simulations are feasible.

Solution with Deposition and Settling

Ermak (1977) derived a modified GP solution:

- Replaces the zero vertical flux BC at $z = 0$ with

$$K_z \frac{\partial C}{\partial z} + W_{set}\, C = W_{dep} C \qquad \text{(deposition flux, } kg/m^2 s\text{)}$$

- Transform as before, but eliminate extra vertical convection term $2w\partial B/\partial Z$ using the substitution

$$B(X,Z) = \overline{B}(X,Z)\,\exp\left[-w(Z-1) - w^2 X\right] \quad \text{with } w = HW_{set}/2K_z.$$

- Yields modified diffusion problems:

$$\frac{\partial A}{\partial X} = \frac{\partial^2 A}{\partial Y^2}$$
$$A(0,Y) = C_o \delta(Y)$$
$$A(X, \pm\infty) = 0$$

$$\frac{\partial \overline{B}}{\partial X} = \frac{\partial^2 \overline{B}}{\partial Z^2}$$
$$\overline{B}(0,Z) = C_o \delta(Z-1)$$
$$\frac{\partial \overline{B}}{\partial Z}(X,0) = \beta \overline{B}(X,0), \quad \overline{B}(X,\infty) = 0$$

Solution with Deposition and Settling

Using Laplace transforms, the solution is:

$$C(x,y,z) = \frac{Q}{2\pi U\sigma_y\sigma_z} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \exp\left(-\frac{W_{set}\,(z-H)}{2K_z} - \frac{W_{set}^2\,\sigma_z^2}{8K_z^2}\right)$$
$$\times \left[\exp\left(-\frac{(z-H)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+H)^2}{2\sigma_z^2}\right) - \sqrt{2\pi}\,\frac{W_o\,\sigma_z}{K_z}\,\exp\left(\frac{W_o(z+H)}{K_z} + \frac{W_o^2\,\sigma_z^2}{2K_z^2}\right) \operatorname{erfc}\left(\frac{W_o\,\sigma_z}{\sqrt{2}\,K_z} + \frac{z+H}{\sqrt{2}\,\sigma_z}\right)\right]$$

where $W_o = W_{dep} - \frac{1}{2}W_{set}$ and $W_{set} = \rho g d^2/18\mu$ (Stokes' law)

Key: Both concentration and deposition flux $(W_{dep}C)$ are linear in Q!!

Other Generalizations

This analytical solution can be modified for a wide range of other situations:

- Line and area sources (Chrysikopoulos et al., 1992).
- Instantaneous or "Gaussian puff" releases (basis for EPA's CALPUFF code).
- Inversion layers introduce a reflecting BC at $z = H_{inv} > H$, leading to a series solution.
- Include vertical dependence, $U(z)$ and $\sigma(z)$, owing to boundary layer structure (Lin & Hildemann, 1996).
- Take $K_x > 0$ to handle $U = 0$ – introduces integral terms (Llewelyn, 1983).

. . . this is a real special function bonanza!!

Other Applications

Stack emissions aren't the only application of plume models:

- Ash from volcanic eruptions (Turner & Hurst, 2001).
- Release of radionucleotides from atomic power plants and weapons blasts (Jeong et al., 1995).
- Biological contaminants: e.g., anthrax release from Sverdlovsk in 1979 (Meselson et al., 1994); terrorist attacks in urban settings with complex geometries.
- Seed dispersal (Levin et al., 2003).
- Insect infestations: locusts, mountain pine beetles.
- Odor propagation from livestock (Chastain & Wolak, 1999).
- Dust and exhaust from automobiles – roads are line sources.

What's left?

So far, we've considered a single source in a constant wind.

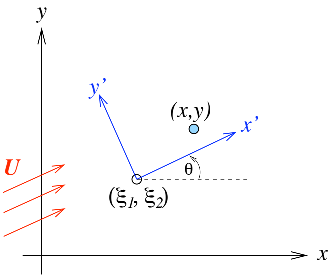
We still need to include:

- Multiple sources (still with constant emission rate).
- Time-dependent wind velocity, not aligned with x-axis.
- Multiple contaminants.
- Ultimately . . . solve the inverse problem.

Time-dependent Wind

For source with location $\vec{\xi}$, and wind with speed $U(t)$ and direction $\theta(t)$:

- Shift and rotate coordinates using $\vec{x}' = R_{-\theta}(\vec{x} - \vec{\xi})$, where R is a rotation matrix.
- Deposition flux is $W_{dep}C(\vec{x}')$.
- Take wind measurements $\{U^n, \theta^n\}$ at times $t^n = n\Delta t$.

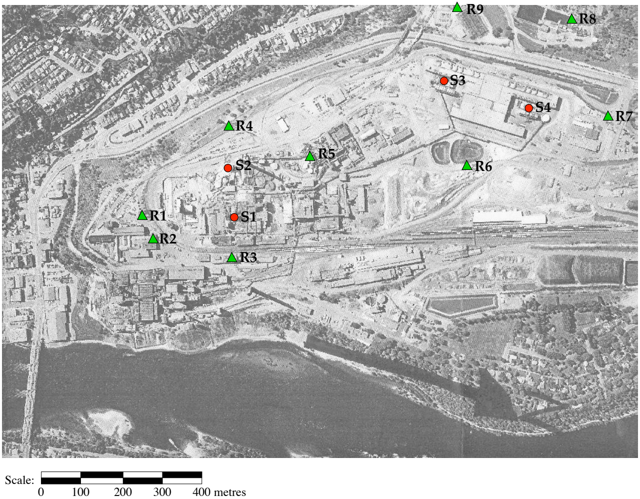


- Rewrite deposition flux as $W_{dep}Q\; p(\vec{x}; \vec{\xi}, U^n, \theta^n)$.
- Total mass deposited within a small cell of area A, centered on $\vec{x}$, over total time $N\Delta t$, is

$$D(\vec{x}) \approx \sum_{n=1}^{N} (W_{dep}QA\Delta t)\; p(\vec{x}; \vec{\xi}, U^n, \theta^n).$$

Multiple Sources

Four sources (Sn) and nine "dustfall jars" or receptors (Rn):



Multiple Sources 2

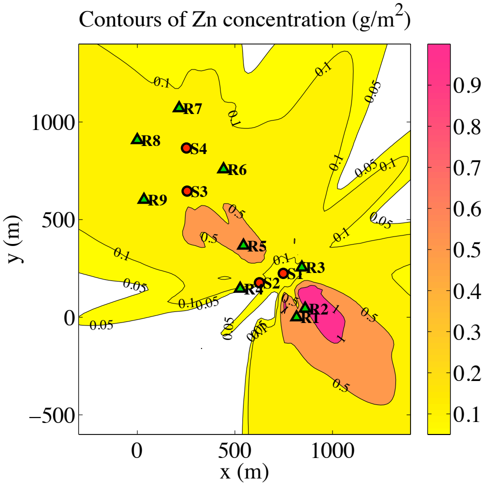
- Sources Q_s have locations $\vec{\xi}_s$ for $s = 1, 2, 3, 4$.
- At any location $\vec{x}$, the deposition from all 4 sources is

$$D_{tot}(\vec{x}) = \sum_{s=1}^{4} \sum_{n=1}^{N} (W_{dep} Q_s A \Delta t)\; p(\vec{x}; \vec{\xi}_s, U^n, \theta^n)$$

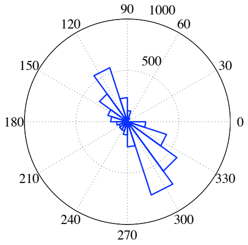
Example: Four sources with different emission rates

Sample Output

One month cumulative zinc deposition using actual wind data:



Wind histogram



Due to Columbia River valley, wind is unidirectional!

Outline

Inverse Problem

- At each receptor location $\vec{\eta}_r$, for $r = 1, 2, \ldots, 9$, measure zinc deposition D_r^{Zn} over one month:

$$D_r^{Zn} = \sum_{s=1}^{4} \sum_{n=1}^{N} (W_{dep} Q_s^{Zn} A \Delta t)\ p(\vec{\eta}_r; \vec{\xi}_s, U^n, \theta^n)$$

  Yields 9 linear equations in 4 unknown Q_s^{Zn} values.
- Obtain a similar system for each $q =$ Zn, Sr, SO_4:

$$P^q\, \vec{Q}^q = \vec{D}^q$$

  where each P^q is 9×4, $\vec{Q}^q$ is 4×1, $\vec{D}^q$ is 9×1.
- Structure: a block diag. system of 27 eqns in 12 unknowns:

$$\begin{bmatrix} P^{Zn} & 0 & 0 \\ 0 & P^{Sr} & 0 \\ 0 & 0 & P^{SO_4} \end{bmatrix} \begin{bmatrix} \vec{Q}^{Zn} \\ \vec{Q}^{Sr} \\ \vec{Q}^{SO_4} \end{bmatrix} = \begin{bmatrix} \vec{D}^{Zn} \\ \vec{D}^{Sr} \\ \vec{D}^{SO_4} \end{bmatrix}$$

Constraints

A number of other physical considerations impose equality and inequality constraints:

- Each emission rate is positive (12 inequalities).
- Molar ratio of Zn relative to SO_4 generated at S1 (1).
- Molar ratio of Zn relative to Sr from cooling towers at S3/S4 (2).
- Sr is only emitted from cooling towers (2).
- Two cooling tower sources are identical (3).

$\Longrightarrow$ An overdetermined linear system with linear constraints (8 equality and 12 inequality)

Ill-conditioning

- The ill-conditioned nature of the unconstrained inverse problem is well-studied in a series of papers by Enting & Newsam (1988-2002).
- When measurements are taken from long distances downwind, or at high altitudes, then the ill-conditioning is severe.
- "The relatively mild degree of ill-posedness in the surface source-deduction problem makes the numerical inversions feasible."

Other Approaches

- Brown (1993): used a finite difference approximation for the forward problem.
- Mulholland & Seinfeld (1995): used a Kalman filtering approach to estimate time-varying sources.
- Bagtzoglou & Baun (2005): solved backwards advection-diffusion equation, using an "equivalent" beam equation that is well-posed.
- Hogan et al. (2005): calculated emission rate and source location (4 variables) using 4 deposition measurements — an idealized case with synthetic data (exact solution!).
- Jeong et al. (1995): used least squares to determine a single emission rate from 51 measurements (very accurate).
- MacKay et al. (2006): nonlinear least squares method for estimating K and W_{dep} for synthetic deposition data and known emission rate (idealized).

Outline

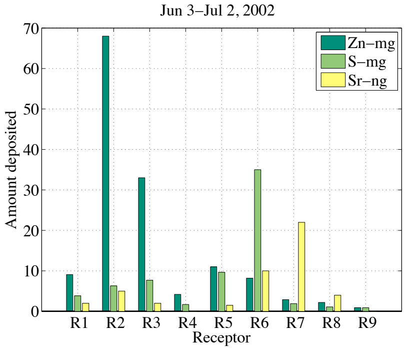
Numerical Simulations

- Use Matlab's constrained linear least squares solver `lsqlin`.
- Each run requires approx. 30 sec. on a Mac laptop – fast!
- Physical parameters were taken from the published literature.
- Sensitivity study: results are most sensitive to
  - stack and receptor heights,
  - atmospheric stability class $\Longrightarrow$ determines $\sigma_{y,z}(x)$,

  and (surprisingly) not so sensitive to noise (even up to 20%).
- Use "engineering estimates" of zinc emissions as a guide:

$$\vec{Q}^{Zn} \approx [35,\ 80,\ 5,\ 5] \ \text{tons/yr}$$

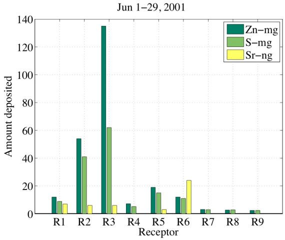
Typical Deposition Measurements

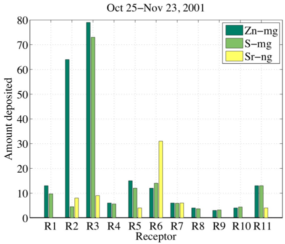


- Most Zn measurements are consistent from month to month.
- Certain Zn measurements exhibit large deviations at R3.
- More variation in SO_4 and Sr data (but less sensitive).

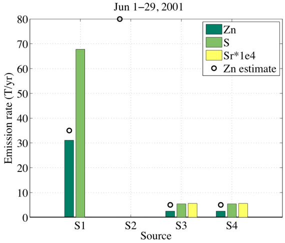
Results

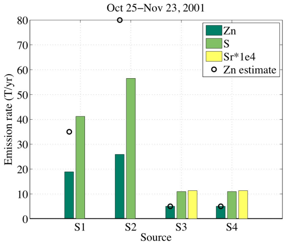
Measured depositions





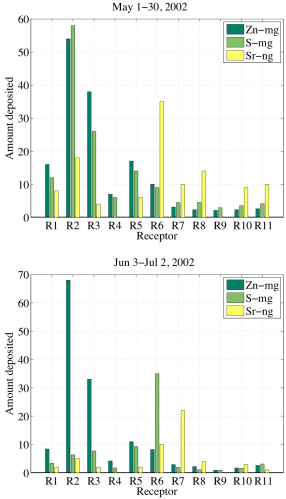
Computed emission rates



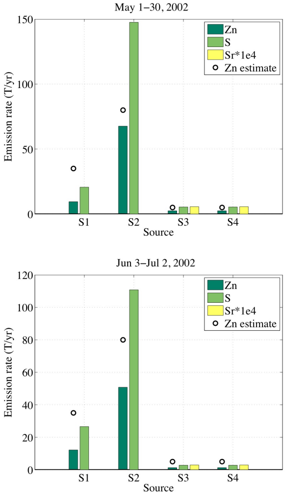


Results 2

Measured depositions



Computed emission rates



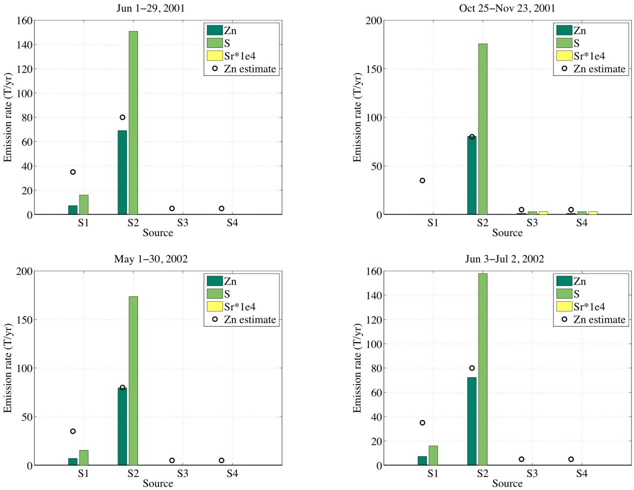
Conclusions

- The method does a reasonable job of capturing total Zn emissions.
- Individual Zn source estimates still vary considerably.
- Assuming near-constant emissions, we'd expect all deposition measurements to be similar

$\Longrightarrow$ suggests that discrepancies in the inverse solution can be attributed to measurement errors (particularly at R3)

Results Without R3

Estimated emissions without R3 are much better (except S3/S4):



Summary

- Used convection-diffusion equation with flux boundary conditions to model contaminant transport.
- Solved exactly using Laplace transforms.
- Linearity (in Q) allowed superposition of sources.
- Given deposition data leads to overdetermined linear system.
- Emissions rates obtained using linear least squares method.
- Ill-posedness limits accuracy of results.
- Total emissions are still reasonable ... which is all that's needed from a regulatory standpoint!
- These results have appeared as

  E. Lushi & JMS, *Atmospheric Environment*, 44(8):1097–1107, 2010.

  and a second paper is soon to be submitted to *SIAM Review*'s "Education" column.

Future Work

- Investigate accuracy of R3 deposition measurements with Teck-Cominco engineers.
- Further validate the algorithm using other deposition measurements for months with missing wind data.
- Relate features of inverse solution to eigenvectors of P matrix (Jackson, 1972).
- Teck-Cominco is currently undertaking another round of deposition measurements (Feb.–Nov. 2010) . . .

Sudeshna Ghosh

- PhD student
- Internship funded jointly by MITACS and Teck-Cominco

On-going Work

We aim to validate some of these results with direct simulations of the advection-diffusion equation:

$$\frac{\partial C}{\partial t} + \vec{u} \cdot \nabla C = \nabla \cdot (K \nabla C) + Q$$

- Use CLAWPACK's high resolution schemes for advective transport.
- Handle diffusion using the Peaceman-Rachford ADI scheme.
- Approximate the delta functions in source terms using

$$\delta_\epsilon(x) = \frac{\epsilon}{\pi(x^2 + \epsilon^2)}$$

- Eddy diffusivities: $K_x = K_y = 2 - 3\ m^2/s$, and K_z taken from Lettau & Dabberdt (1970):

$$K_z(z) = 0.6033 + 0.0185z - 0.000108z^2\ m^2/s$$

Thank-you!

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