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Method development estimating atmospheric deposition of various pollutants

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Contents

- A bulk particle dry deposition algorithm
- A size-resolved and bulk algorithm for precipitation scavenging of aerosol particles
- A modified gradient method for dry deposition estimation over forests
- A framework estimating atmospheric deposition in the Canadian oil sands region

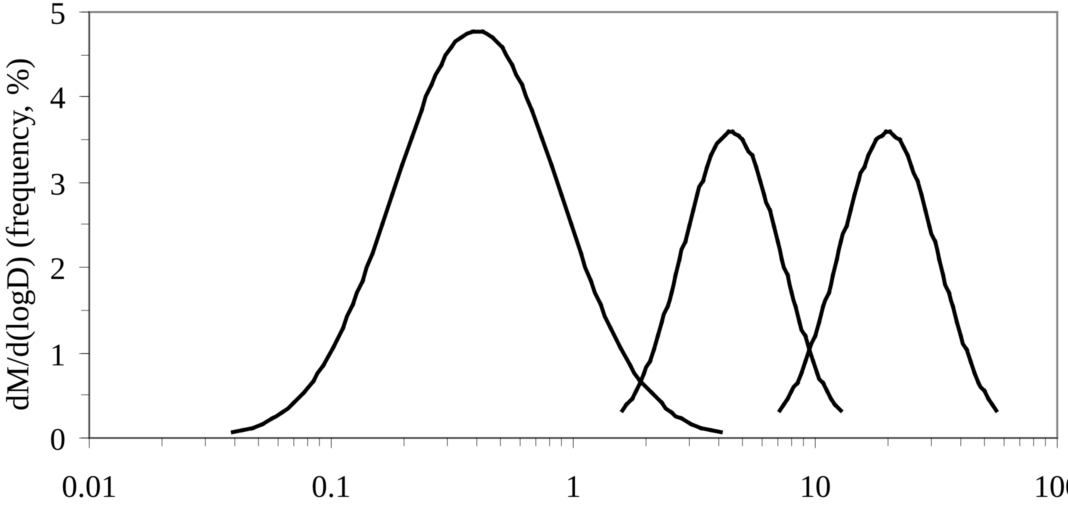
Development of a bulk dry deposition algorithm for fine, coarse and giant particles

- **Motivation**
  - Monitoring network only collect bulk aerosols
  - Some CTMs use modal approach or coarse size-bin
- **Findings and recommendations from Zhang et al. (2012, ACP)**
  - Particle dry deposition can be estimated in reasonable accuracy if the mass factions of $PM_{2.5}$, $PM_{2.5-10}$, and PM_{10+} are know.
  - Empirical deposition algorithms should not be 'particle-species'-dependant; they should be for certain particle size ranges

Development of a bulk dry deposition algorithm for fine, coarse and giant particles

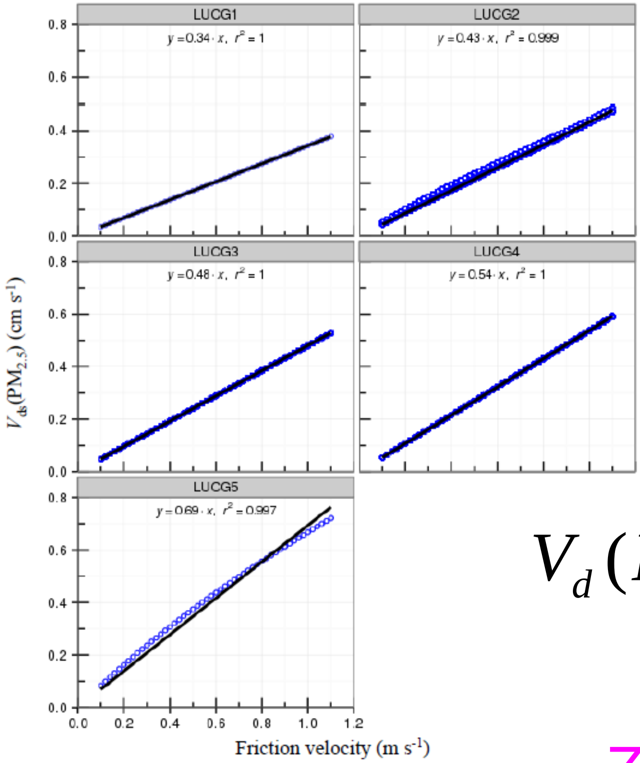
- **Taking Zhang et al. (2001) as a benchmark model**
  - The most widely used scheme
  - Applicable to any surface types
  - Ability to predict reasonable deposition velocity (Vd)
- **Generate a Vds (=1/Rs) database using assumed size distribution**

$$V_d = V_g + \frac{1}{R_a + R_s}$$



Development of a bulk dry deposition algorithm for fine, coarse and giant particles

- **Develop the algorithm from the newly-generated database**

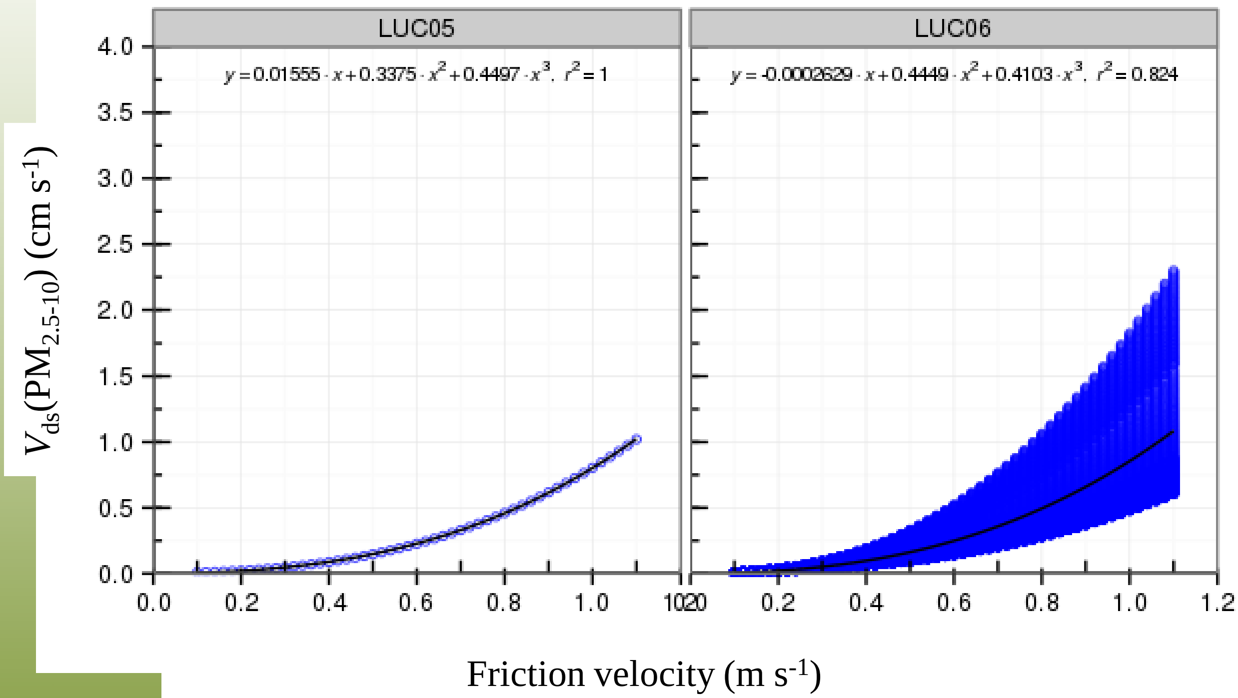


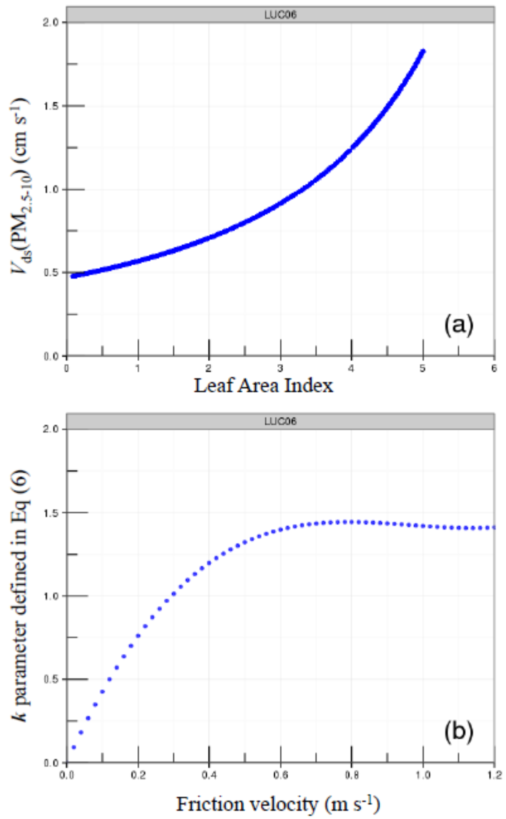
$$V_{ds} = a_1 u_*$$

$$V_d(PM_{2.5}) = V_g(PM_{2.5}) + \frac{1}{R_a + 1/(a_1 u_*)}$$

Zhang and He (2014, ACP)

Development of a bulk dry deposition algorithm for fine, coarse and giant particles





$$V_{ds}(PM_{2.5-10}) = b_1 u_* + b_2 u_*^2 + b_3 u_*^3$$

$$V_{ds}(PM_{2.5-10}) = (b_1 u_* + b_2 u_*^2 + b_3 u_*^3) e^{(c_1 u_* + c_2 u_*^2 + c_3 u_*^3)(\frac{LAI}{LAI_{max}} - 1)}$$

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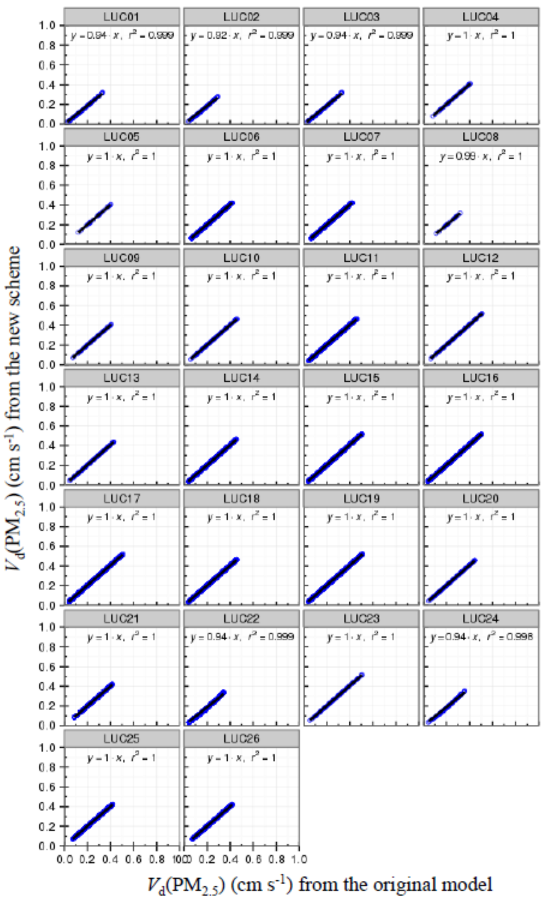
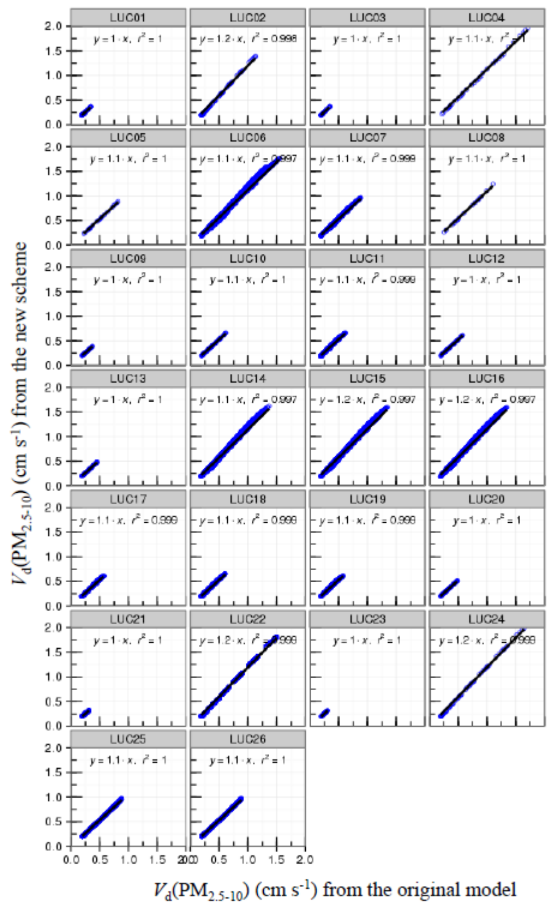
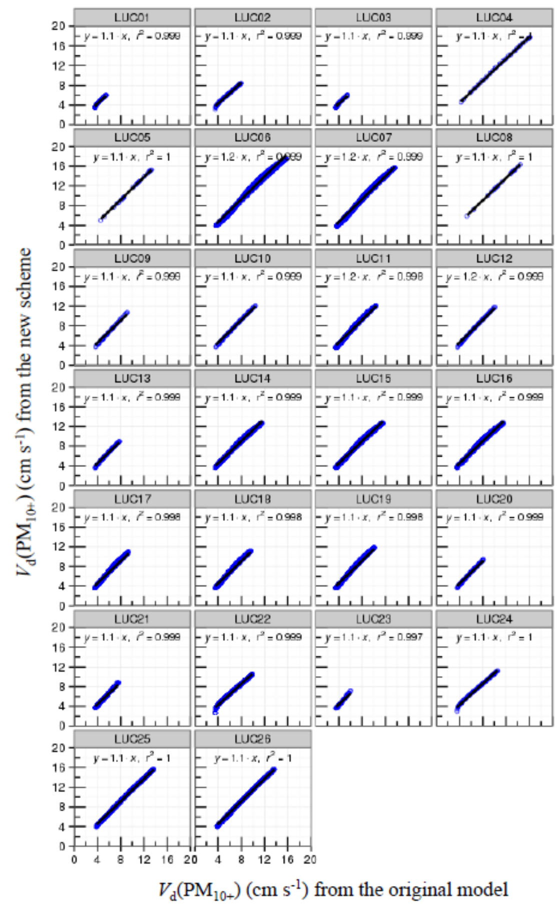
Development of a bulk dry deposition algorithm for fine, coarse and giant particles

- **Comparison between the new and old scheme**







Zhang and He (2014, ACP)

Development of a bulk dry deposition algorithm for fine, coarse and giant particles

- **Example application to AMNet**

$$F_{PBM} = C_{PBM} * V_{d\,PBM}$$

$$C_2 = C_{PBM} \frac{f}{1-f}$$

f -mass fraction of coarse PBM

$$F_{TPBM} = C_{PBM} V_{d\,PBM} + C_2 V_{d2} = C_{PBM} V_{d\,PBM} + C_{PBM} \frac{f}{1-f} V_{d2} = F_{PBM} \left(1 + \frac{f}{1-f} \frac{V_{d2}}{V_{dPBM}}\right)$$

Flux missed by the network

Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

The scavenging coefficient Λ (s^{-1}) describes the scavenging rate or the fractional reduction per unit time of either bulk or size-resolved aerosol particle number or mass concentration

Bulk: $$\frac{\partial n(t)}{\partial t} = -\Lambda \cdot n(t)$$

Size-resolved: $$\frac{\partial n(d_p, t)}{\partial t} = -\Lambda(d_p) \cdot n(d_p, t)$$

$$\Lambda(d_p) = \int_0^\infty \frac{\pi}{4}(D_p + d_p)^2 \left(V(D_p) - v(d_p)\right) E(d_p, D_p) N(D_p) dD_p$$

D_p and d_p are hydrometeor and aerosol-particle diameter,

V and v are hydrometeor and aerosol-particle fall velocity,

$E(d_p, D_p)$ is hydrometeor-aerosol particle collection efficiency,

$N(D_p)$ is hydrometeor number size distribution.

Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

Contributing Studies

Wang X., Zhang L., and Moran M.D., 2010: **Uncertainty assessment of current size-resolved parameterizations for below-cloud particle scavenging by rain**. *Atmos. Chem. Phys.*, 10, 5685-5705.

Wang X., Zhang L., and Moran M.D., 2011. **On the discrepancies between theoretical and measured below-cloud particle scavenging coefficients for rain - a numerical investigation using a detailed one-dimensional cloud microphysics model**. *Atmos. Chem. Phys.*, 11, 11859-11866.

Zhang L., Wang X., Moran M.D., and Feng J., 2013. **Review and uncertainty assessment of size-resolved scavenging coefficient formulations for snow scavenging of atmospheric aerosols**. *Atmos. Chem. Phys.*, 13, 10005-10025.

Wang X., Zhang L., and Moran M.D., 2014. **Development of a new semi-empirical parameterization for below-cloud scavenging of size-resolved aerosol particles by both rain and snow**. *Geoscientific Model Development*, 7, 799–819.

Wang X., Zhang L., and Moran M.D., 2014. **A bulk parameterization for below-cloud scavenging of fine, coarse and giant particles by both rain and snow**. *Journal of Advances in Modeling Earth Systems*, in press.

Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

- **Recommendations from review and uncertainty assessment studies**

- Empirical formulas should not be used in CTMs because some of the processes contributing to the field-derived estimates are treated in CTMs separately
- Upper-range values of available theoretical formulations should be used in CTMs because they are closer to, while still smaller than, the field-derived estimates
- A simple semi-empirical formula for size-resolved scavenging coefficient should be developed that takes into account the large range of its values that can be obtained from existing theoretical formulas, the many different possible choices for their product terms, and the upper-bound values provided by field-derived estimates.

Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

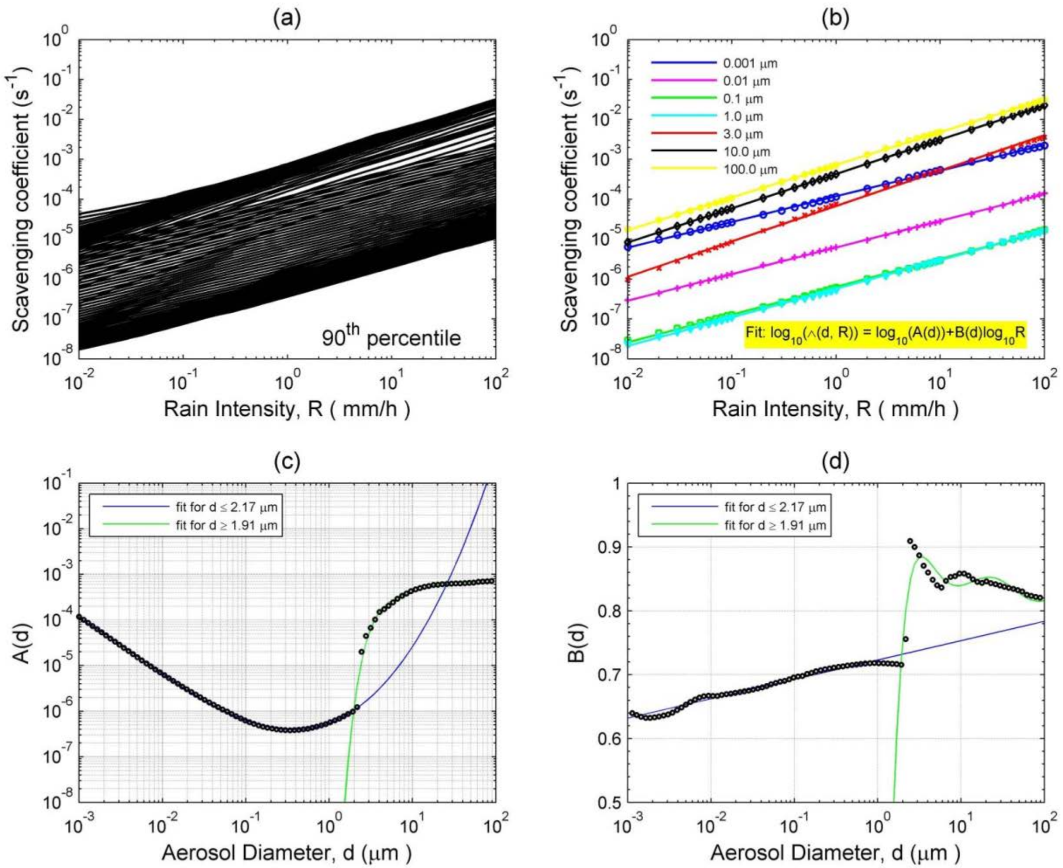
- **Methodology for Λ parameterization development values**

- Develop an ensemble of theoretical parameterizations for $\Lambda(d_p)$ based on consideration of all available formulas for $E(d_p, D_p)$, $N(D_p)$, $V(D_m)$, and $A(D_m)$ for rain and for snow

- For each value of d_p, generate a database using the 90th values of $\Lambda(d_p)$ for selected precipitation intensities R from 0.01 to 100 mm h^{-1} for rain and from 0.001 to 10 mm h^{-1} for snow (as liquid water equivalent)

Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

- **Develop semi-empirical formulas based on the new database**



Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

$$\log_{10}(\Lambda(d,R)) = \log_{10}(A(d)) + B(d)(\log_{10} R)$$

$$\Lambda(d,R) = A(d)R^{B(d)}$$

For rain scavenging

$$\log_{10}(A(d)) = \begin{cases} a_0 + a_1(\log_{10} d) + a_2(\log_{10} d)^2 + a_3(\log_{10} d)^3 & d \le 2.0\mu m \\ b_0 + b_1(\log_{10} d) + b_2(\log_{10} d)^2 + b_3(\log_{10} d)^3 + b_4(\log_{10} d)^4 + b_5(\log_{10} d)^5 + b_6(\log_{10} d)^6 & d > 2.0\mu m \end{cases}$$

$$B(d) = \begin{cases} c_0 + c_1(\log_{10} d) & d \le 2.0\mu m \\ e_0 + e_1(\log_{10} d) + e_2(\log_{10} d)^2 + e_3(\log_{10} d)^3 + e_4(\log_{10} d)^4 + e_5(\log_{10} d)^5 + e_6(\log_{10} d)^6 & d > 2.0\mu m \end{cases}$$

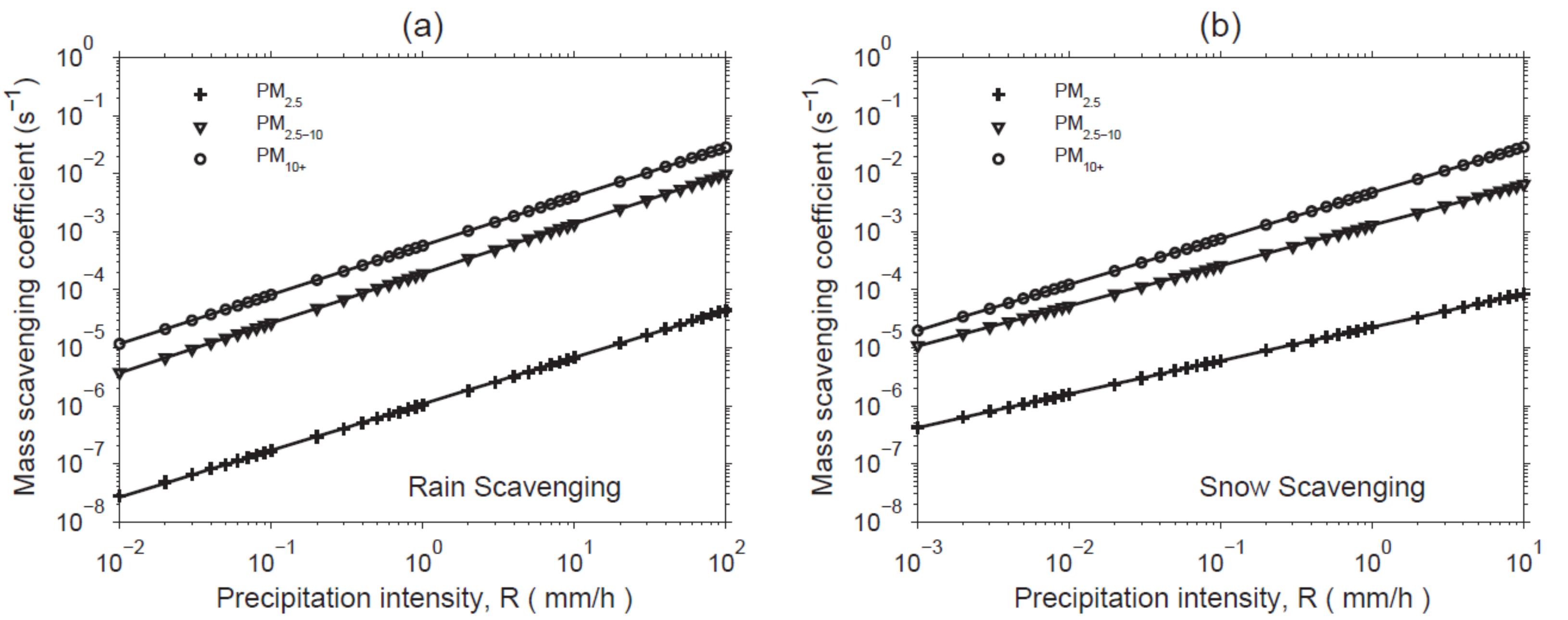
For snow scavenging

$$\log_{10}(A(d)) = \begin{cases} a_0 + a_1(\log_{10} d) + a_2(\log_{10} d)^2 + a_3(\log_{10} d)^3 + a_4(\log_{10} d)^4 + a_5(\log_{10} d)^5 + a_6(\log_{10} d)^6 & d \le 1.44\mu m \\ b_0 + b_1(\log_{10} d) + b_2(\log_{10} d)^2 + b_3(\log_{10} d)^3 + b_4(\log_{10} d)^4 + b_5(\log_{10} d)^5 + b_6(\log_{10} d)^6 & d > 1.44\mu m \end{cases}$$

$$B(d) = \begin{cases} c_0 + c_1(\log_{10} d) + c_2(\log_{10} d)^2 + c_3(\log_{10} d)^3 + c_4(\log_{10} d)^4 + c_5(\log_{10} d)^5 + c_6(\log_{10} d)^6 & d \le 1.44\mu m \\ e_0 + e_1(\log_{10} d) + e_2(\log_{10} d)^2 + e_3(\log_{10} d)^3 + e_4(\log_{10} d)^4 + e_5(\log_{10} d)^5 + e_6(\log_{10} d)^6 & d > 1.44\mu m \end{cases}$$

Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

- **Extension of the algorithm to bulk scavenging – for applications in modal-approach or coarse-bin CTMs**



Development of a size-resolved and bulk algorithm for precipitation scavenging of aerosols

$$\log_{10}(\Lambda_m) = \log_{10} A + B(\log_{10} R) \quad \text{or} \quad \Lambda_m = AR^B$$

		$PM_{2.5}$	$PM_{2.5\text{-}10}$	PM_{10+}
$\Lambda_{m,rain}$	**A**	1.065×10^{-6}	1.896×10^{-4}	5.753×10^{-4}
	B	8.022×10^{-1}	8.558×10^{-1}	8.476×10^{-1}
	r^2	9.999×10^{-1}	1.0	1.0
$\Lambda_{m,snow}$	**A**	2.241×10^{-5}	1.300×10^{-3}	4.700×10^{-3}
	B	5.762×10^{-1}	6.957×10^{-1}	7.915×10^{-1}
	r^2	1.0	1.0	1.0

Development of a modified gradient method for flux estimation over forest canopies

- **Existing micrometeorological methods**
  - **Eddy-covariance technique (EC)**

$$F = \overline{w'c'}$$

  - **Modified Bowen Ratio method (MB)**

$$F = F_{co_2} \Delta C(O_3) / \Delta C(CO_2)$$

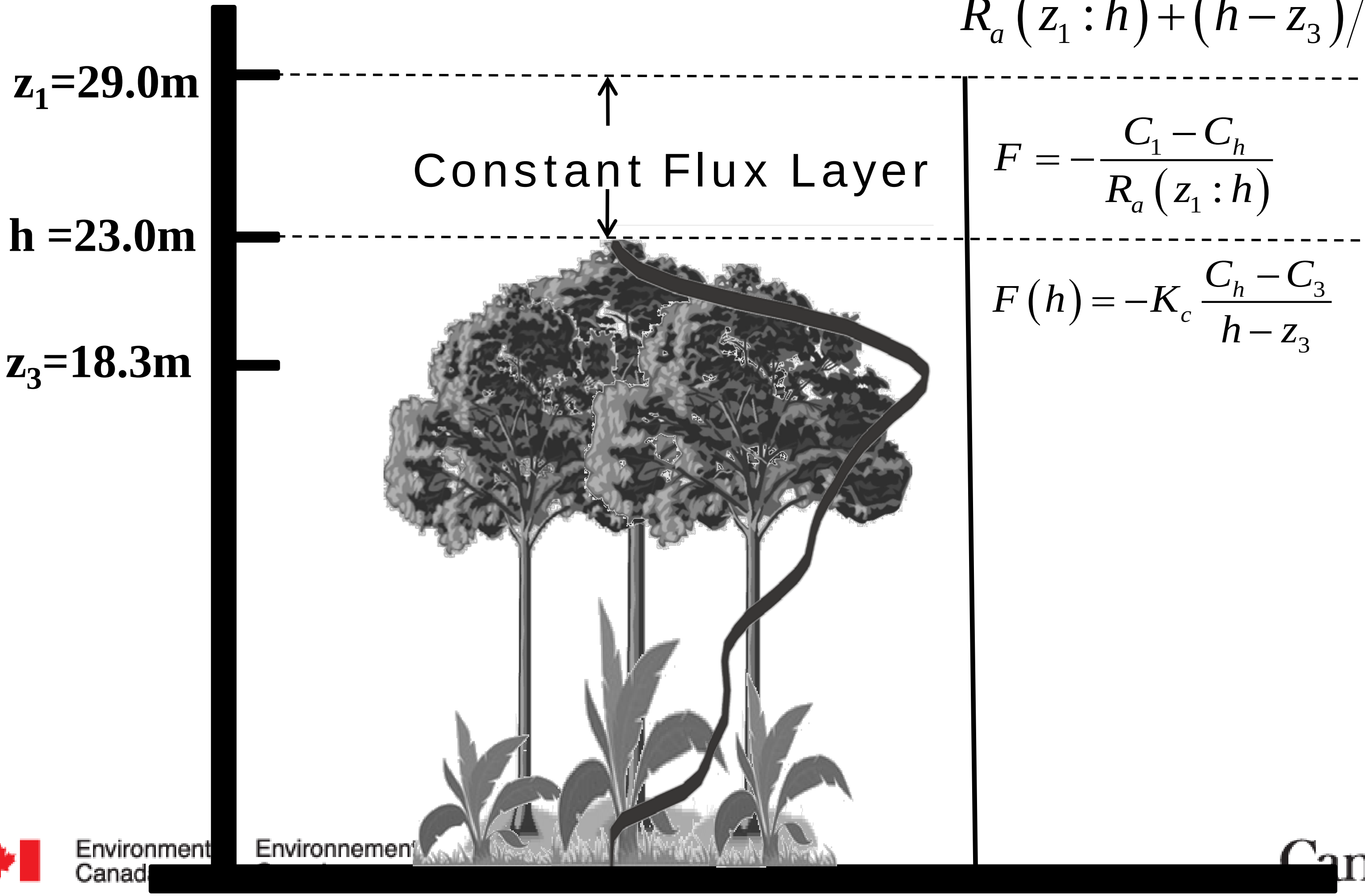
  - **Aerodynamic Gradient method (AG)**

$$F = -\frac{\Delta C}{R_a(z_1 : z_2)} = -\frac{C_1 - C_2}{R_a(z_1 : z_2)}$$

Development of a modified gradient method for flux estimation over forest canopies

- **Modified gradient method**

$$F = -\frac{C_1 - C_3}{R_a\left(z_1 : h\right) + \left(h - z_3\right)/K_c}$$

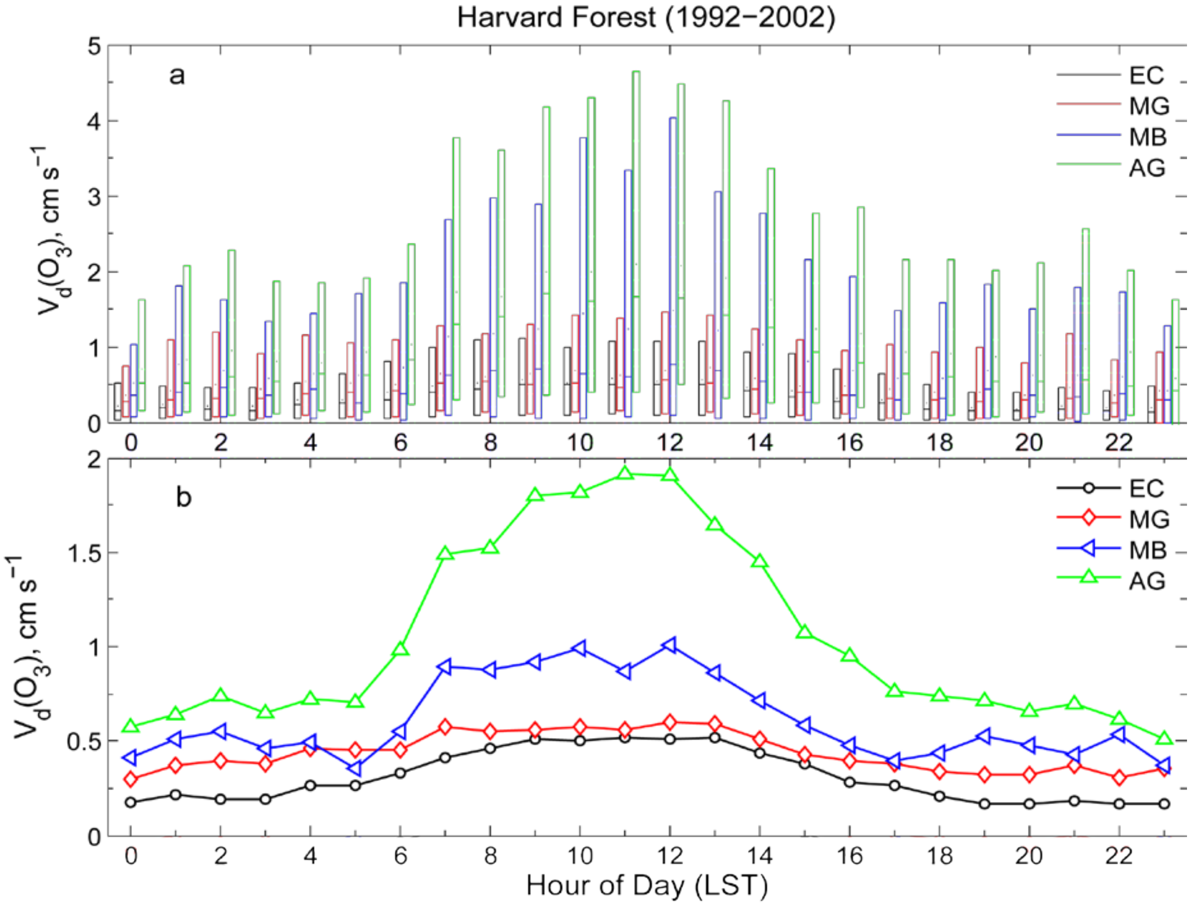


$$F = -\frac{C_1 - C_h}{R_a\left(z_1 : h\right)}$$

$$F\left(h\right) = -K_c \frac{C_h - C_3}{h - z_3}$$

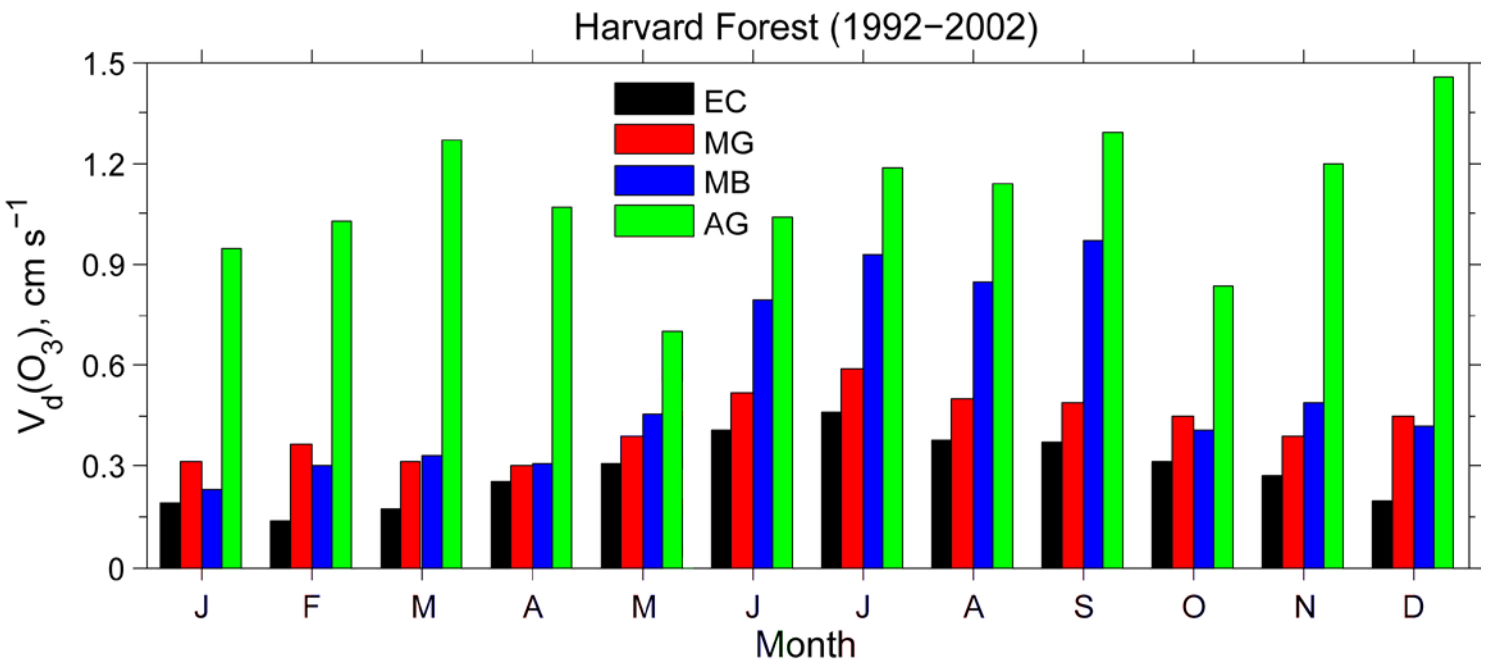
Development of a modified gradient method for flux estimation over forest canopies

- **Diurnal variation of $V_d(O_3)$**



Development of a modified gradient method for flux estimation over forest canopies

➢ **Monthly variation of $V_d(O_3)$**



Development of a framework estimating deposition of pollutants monitored in the Canadian oil sands region

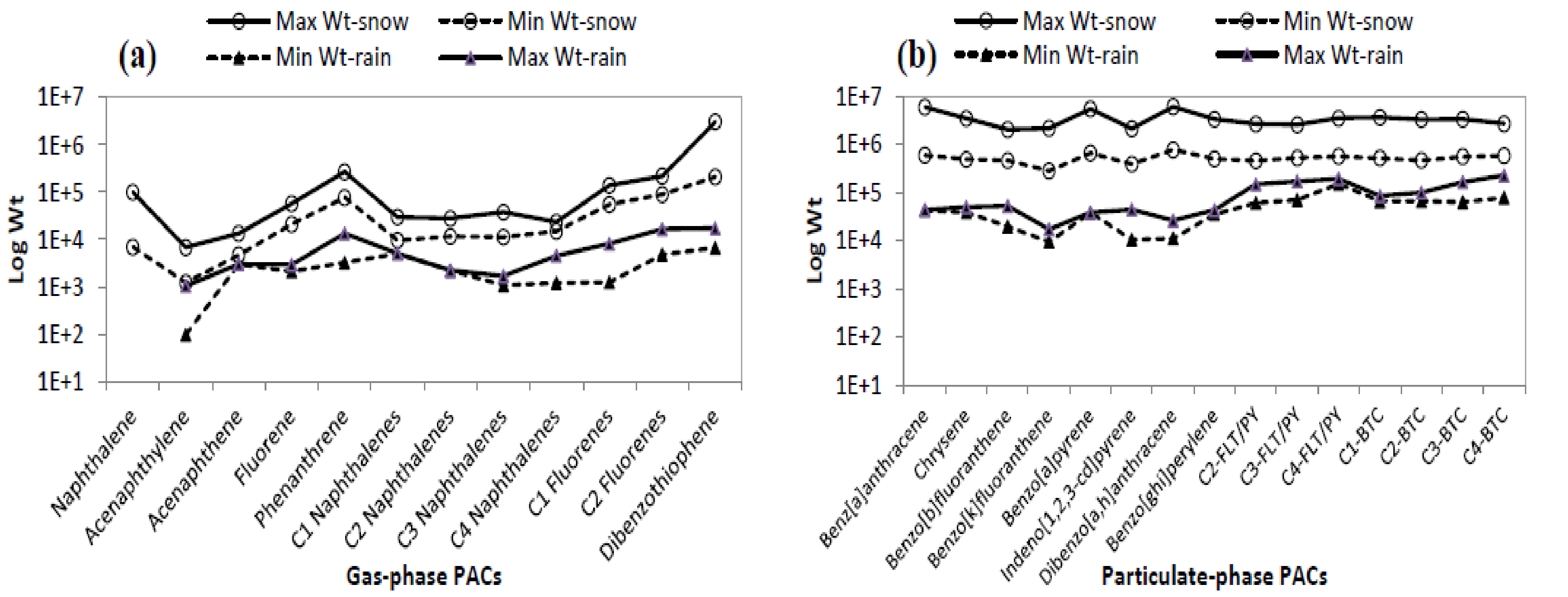
- **Data availability**
  - Polycyclic aromatic compounds (40+ species) : surface air concentration at 3 active sampling and 17 passive sampling sites, and precipitation concentration at 3 sites.
  - Trace elements (40+ species): surface air and precipitation concentration at 3 sampling sites
  - Satellite SO_2 and NO_2 covering the whole region
  - Archived forecasted surface-layer meteorology

Development of a framework estimating deposition of pollutants monitored in the Canadian oil sands region

- **Methodology**
- Dry deposition – inferential method
- Wet deposition – direct measurement and scavenging ratio concept
- A long to-do list on various scientific issues (develop PACs dry deposition scheme, collect size distribution of various PM species, develop/collect scavenging ratio data, develop spatial extrapolation/interpolation method for mapping purpose, etc.

Development of a framework estimating deposition of pollutants monitored in the Canadian oil sands region

➢ **Example progress –scavenging ratios of PACs**

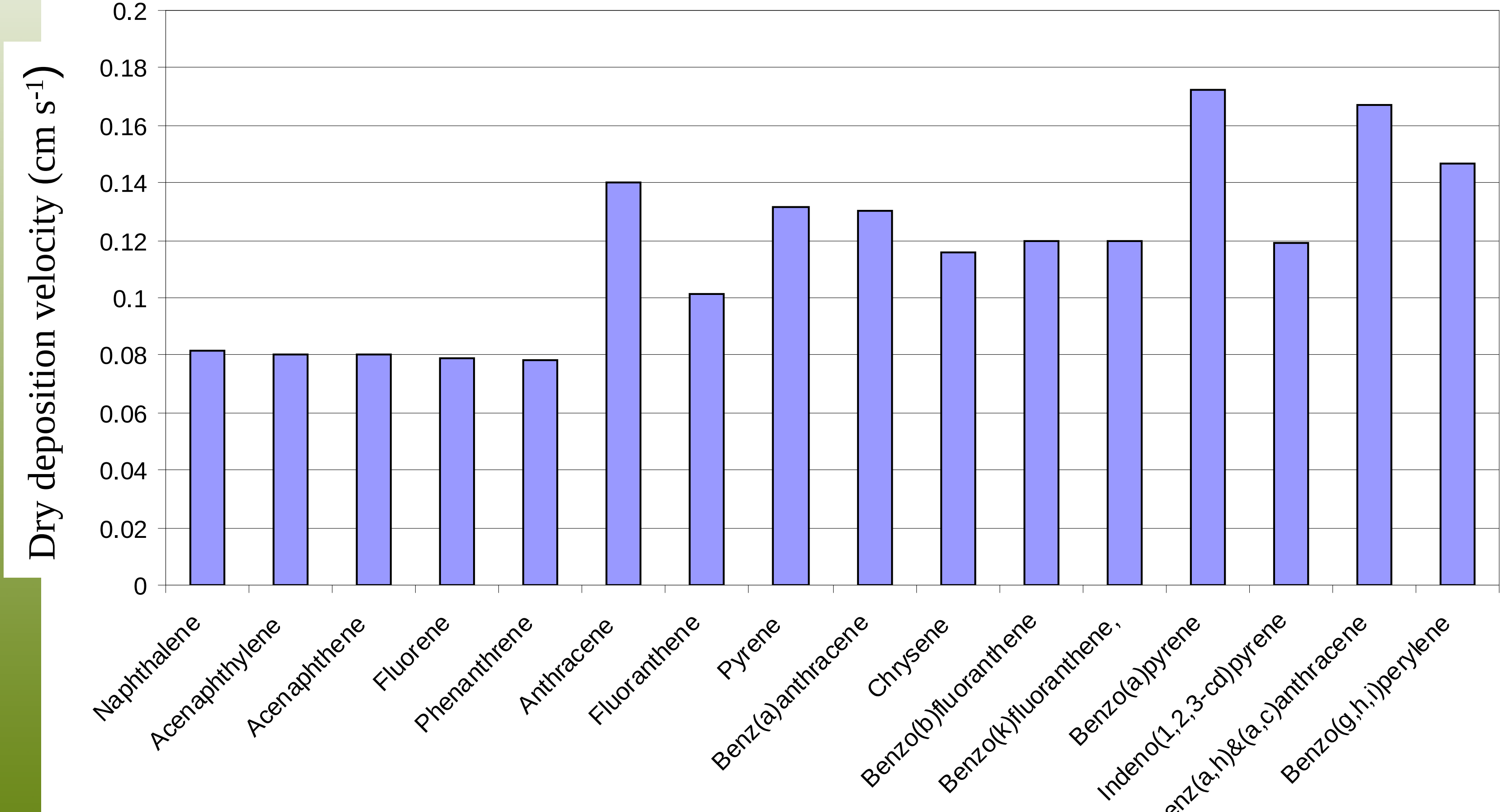


Range of total scavenging ratios (Wt) in snow and rain samples with similar precipitation rates (snow: 11.6-11.8 mm, rain: 10.8-12.3 mm per month)

Zhang et al. (2014, ACP)

Development of a framework estimating deposition of pollutants monitored in the Canadian oil sands region

- **Example progress - dry deposition velocities for gaseous PACs**



For Mannix - LUC: 34% mixed forest, 29% wetland, 37% bare soil

Development of a framework estimating deposition of pollutants monitored in the Canadian oil sands region

- **Example progress - Estimation of particle fraction for PACs**
  - **From the concentration of total suspended particles (TSP) and the PAC's particle/gas partition coefficient K_P**

$$\Phi = \frac{TSP * K_P}{TSP * K_P + 1}$$

$$TSP = m_0 * e^{m_1 * Distance} + 10$$

$$log K_P = log K_{oa} + log f_{om} - 11.91$$