Estimating sources, sinks and fluxes of reactive atmospheric compounds within a forest canopy

Tomer Duman[1], John T. Walker[2], Jesse O. Bash[2], Khaled Ghannam[1], Cheng-Wei Huang[1], Andrey Khlystov[4], Gabriel G. Katul[1,3]

1. Nicholas School of the Environment, Duke University, Durham, North Carolina, USA; 2. US Environmental Protection Agency, Research Triangle Park, North Carolina, USA
3. Department of Civil and Environmental Engineering, Duke University, Durham, North Carolina, USA; 4. Division of Atmospheric Sciences, Desert Research Institute, Reno, Nevada, USA

Introduction

Canopy-scale flux measurements and inferential models are useful for developing estimates of net emission or deposition of trace gases and aerosols above forests. However, more detailed measurements and models are needed to relate net fluxes to biological, physical, and chemical processes occurring within the air-canopy-soil system. These processes occur over multiple time scales making direct measurements of sources or sinks difficult to conduct. However, measured concentration profiles can be used to infer the effective source-sink distribution – commonly referred to as the solution of the **inverse problem**. Methods for estimating sources or sinks of reactive nitrogen within a forest canopy are explored.

Methods

1. Eulerian frame of reference

   Estimating the effective source (S) from the steady state mean scalar conservation equation:

$$-\frac{\mathrm{d}\overline{w'c'}}{\mathrm{d}z} + S = 0$$

   where the flux $\overline{w'c'}$ is estimated from the corresponding flux budget equation:

$$\frac{\mathrm{d}\overline{w'c'}}{\mathrm{d}t} = -\overline{w'w'}\frac{\mathrm{d}C}{\mathrm{d}z} - \frac{\mathrm{d}\overline{w'c'c'}}{\mathrm{d}z} - \overline{c'\frac{\mathrm{d}p'}{\mathrm{d}z}}$$

   with the following closures:

$$\overline{w'c'c'} = \frac{\tau}{C_8}\left[-\overline{w'w'w'}\frac{\mathrm{d}C}{\mathrm{d}z} - \overline{w'c'}\frac{\mathrm{d}\overline{w'w'}}{\mathrm{d}z} - 2\overline{w'w'}\frac{\mathrm{d}\overline{w'c'}}{\mathrm{d}z}\right], \quad \overline{c'\frac{\mathrm{d}p'}{\mathrm{d}z}} = C_4\frac{\overline{w'c'}}{\tau}$$

2. Lagrangian localized near-field theory - LNF

   A dispersion kernel is generated by dividing the canopy to n layers and calculating the concentration profiles for a uniformly distributed unit source from each of these layers – resulting in a transformation matrix (D) that relates S to mean concentration (C):

   $C - C_R = D\,S$ where C_R is a reference concentration

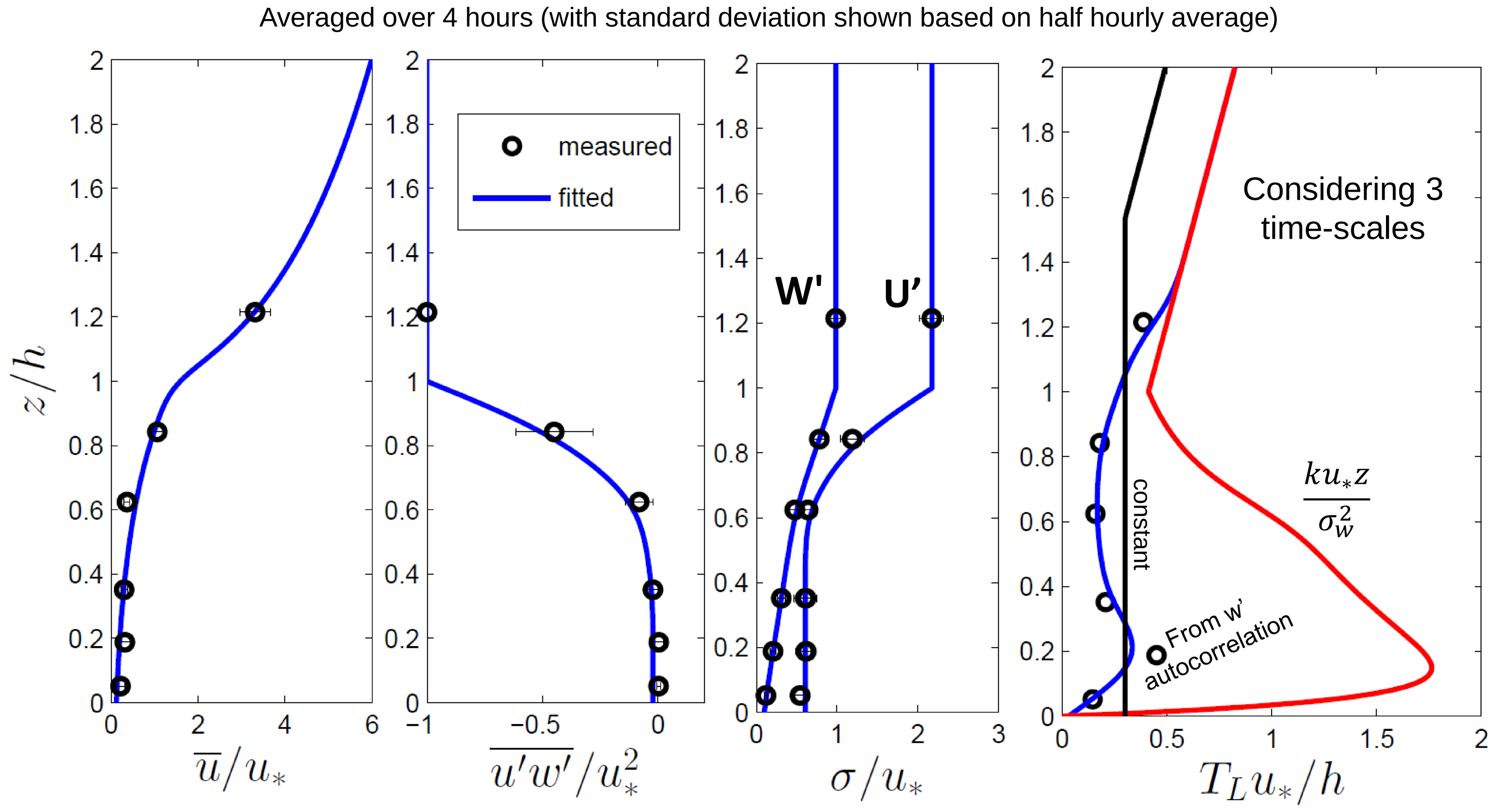
   In the LNF, D is calculated by superimposing near- (C_n) and far- (C_f) field contributions, assuming vertical dispersion, locally homogenous turbulence, and constant time scale.

3. Lagrangian stochastic method

   The D is calculated from a full Lagrangian stochastic scheme, taking into account the covariance between vertical (w') and horizontal (u') velocity components.

Flow statistics used for source estimation

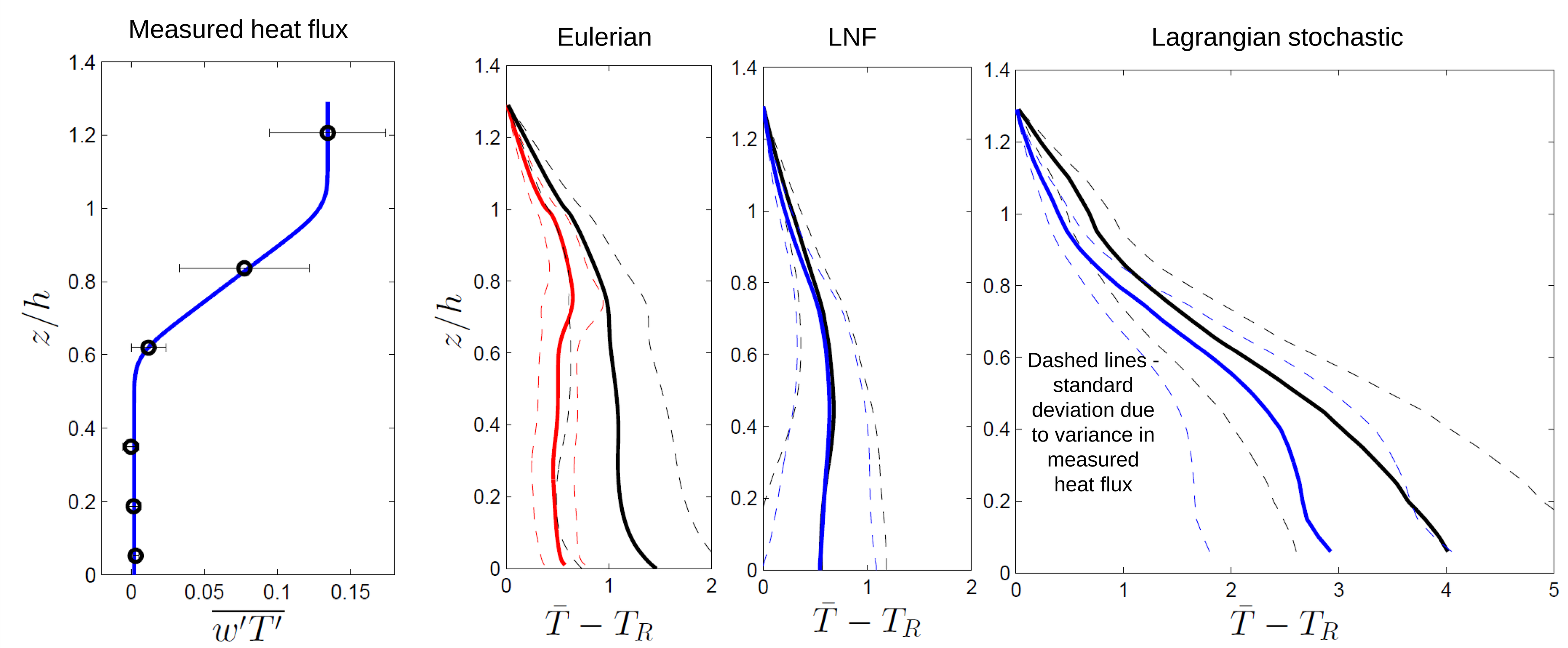
Averaged over 4 hours (with standard deviation shown based on half hourly average)



Statistics include mean wind speed ($\overline{u}$), friction velocity (u_*), velocity variance (σ), and Lagrangian time scale (T_L).

Sensitivity analysis

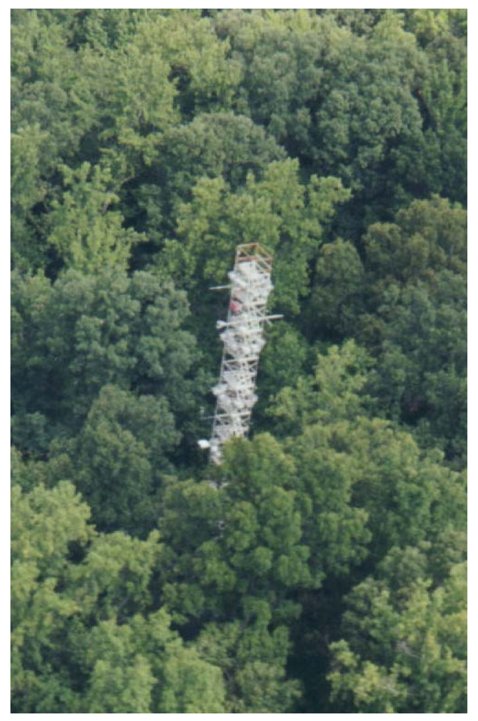
To assess the three methods of source estimation, a measured height (z) resolved heat flux ($\overline{w'T'}$) profile is used to calculate the air temperature profile ($\overline{T}$) relative to the reference temperature (T_R) at canopy height (h).

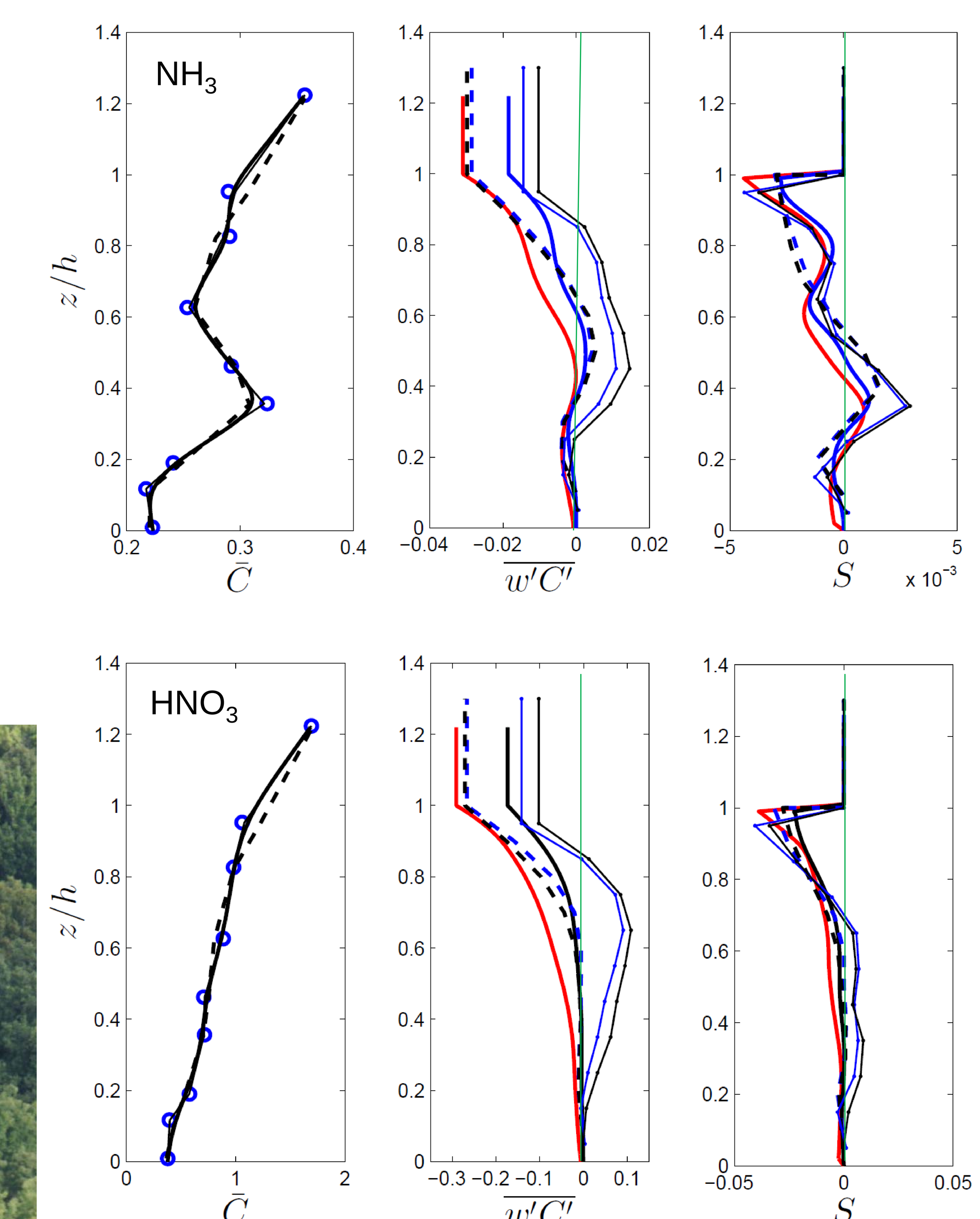


Preliminary results
source/sink estimation of reactive nitrogen within a forest canopy

In-canopy and above-canopy multi-level mean concentration measurements of reactive nitrogen compounds, as well as other compounds that are highly reactive to ammonia (hydrochloric acid and sulfur dioxide) were performed within a deciduous second-growth 180 - year old oak-hickory forest situated within the Southeastern U.S.

Preliminary results (right) are shown for NH_3 and HNO_3 concentrations ($\overline{C}$) measured during September over a 4-hour period. The flux profiles ($\overline{w'C'}$) show sensitivity to the chosen model. However, all models capture similar trends for the source/sink distribution (S) despite differences in underlying assumptions.

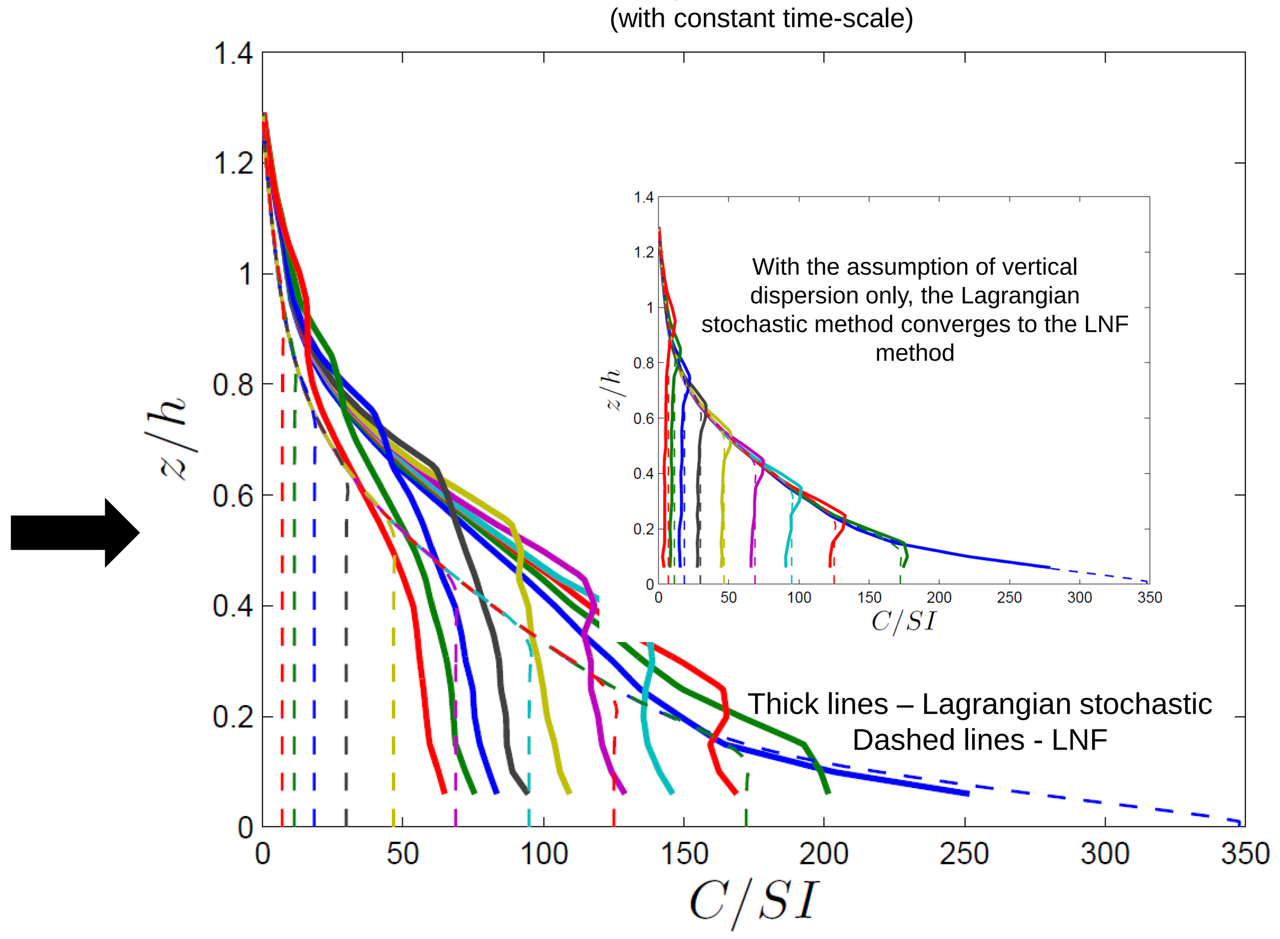




Thick lines – Eulerian model
Dashed lines – LNF
Thin lines – Lagrangian model

Dispersal kernel

(with constant time-scale)



Height resolved concentration (C) normalized by source intensity (SI)

Concentrations of NH_3, HNO_3, SO_2, HONO, and HCl were measured at multiple heights within and above the canopy, using annular denuders (URG Corporation), over collection periods of 3 – 4 hours. Fine wire thermocouples (OMEGA Eng., Inc.) were co-located with denuders. Wind and temperature were measured at a subset of heights using R.M. Young (Model 81000V) sonic anemometers.

Eulerian and LNF models show reasonable profile shapes for HNO_3, for which the canopy and soil should be a sink (negative S), and NH_3, which can be exchanged bi-directionally with vegetation or soil depending on the underlying compensation point.

Future directions

Reactive species undergo turbulent dispersion within an inhomogeneous flow field and ma emission, deposition or transformations on leaves, woody elements, and the forest floor. The quantification of these processes within a source/sink framework is a step toward understanding air pollutant exposure to specific ecological endpoints and feedback to ecological function and atmospheric composition.

The source-sink distribution of the measured compounds can be further estimated from mean concentration measurements at different times with different meteorological conditions, allowing the assessment of the role of dispersion in the evolution of these sources or sinks.

Partitioning of the various processes and their contribution to the source-sink distribution will be achieved by a comparison of different chemical species, as each of these is subjected to different chemical, biological and/or physical processes.

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