Assessment of various background approaches for their use in deriving regional emissions from atmospheric data

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Motivation

With increasing concentrations of greenhouse gases in the atmosphere, efforts are being made to quantify their global and regional emissions. Relative to a global scale, greenhouse gas emissions on a regional scale are less certain and more difficult to quantify. Improvement in regional emission estimates would enable us to evaluate environmental policies associated with controlling greenhouse gas emissions from human activities.

The atmospheric data can be used to derive regional emissions via tracer-tracer correlation or inversion analysis, in some of which it requires estimation on the background or upwind concentrations. The goal of estimating accurate background is so that the derived enhanced concentrations can provide an accurate representation of the associated emission.

Various Approaches of Background Estimates

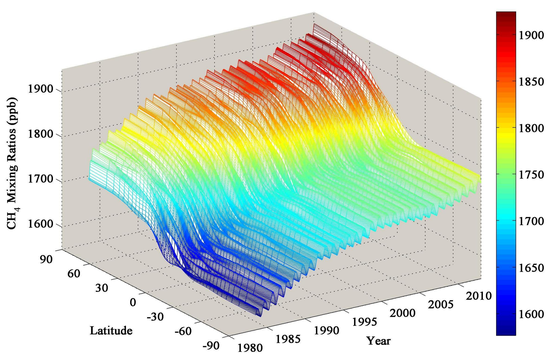


Fig. 1. Global surface background mixing ratios of CH_4 as a function of time and latitude.

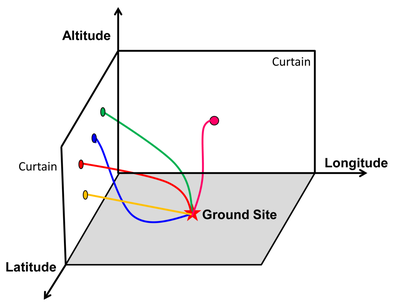


Fig. 2. Schematic diagram of background approach (4). Red star stands for the location of atmospheric measurements; color lines stand for air back-trajectories; "curtain" stands for 2-D background concentrations at the boundaries of a studied region.

Approach (1): Using the lower 5th or 10th percentile of the atmospheric data.

Approach (2): Using the mean/median free tropospheric mixing ratios from aircraft measurements.

Approach (3): Using the surface background mixing ratios at sampling latitudes. Fig. 1 shows an example of global surface background mixing ratios of CH_4, which were derived based on data from remote boundary sites across the globe (See Masarie and Tans (1995) for the detailed method).

Approach (4): Using the Lagrangian particle dispersion model (LPDM) as a tool to sample the concentration of a studied gas at the boundary of a studied region (Fig. 2).

Comparison Between Background Derived from Different Approaches

Experiment 1: We used CO, CH_4, HCFC-22 and HFC-134a as examples to examine the difference in their derived enhanced mixing ratios ($\Delta C = C_{obs} - C_{bkg}$) from various background approaches at West Branch, Iowa (WBI), USA (Fig. 3).

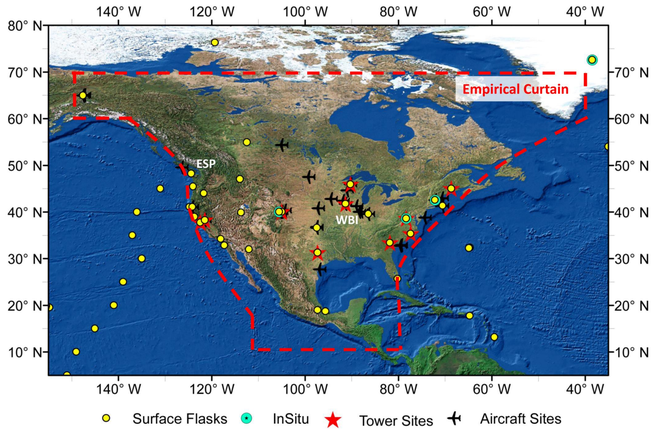


Fig. 3. Map of our North America monitoring sites. The "empirical curtain" stands for 2D background mixing ratios of various gases at the North America boundary. It was constructed using free tropospheric aircraft data from all North America aircraft sites, along with air back-trajectories.

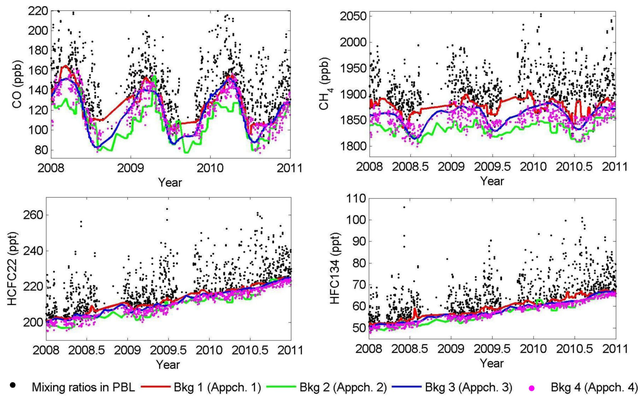


Fig. 4. Observed mixing ratios of CO, CH_4, HCFC-22 and HFC-134a at WBI during 2008 – 2010 and their background derived from four different approaches.

Table 1. Derived enhanced mixing ratios (ΔC) of CO, CH_4, HCFC-22 and HFC-134a using observed and derived background mixing ratios from four different approaches.

	ΔC (mean ± 1 σ)			
	Bkg. Apprch. (1)	Bkg. Apprch. (2)	Bkg. Apprch. (3)	Bkg. Apprch. (4)
CO (ppb)	20.5 (±19.0)	35.4 (±22.3)	27.8 (±21.6)	25.7 (±20.9)
CH_4 (ppb)	35.4 (±33.7)	75.8 (±35.0)	54.4 (±35.5)	60.1 (±38.5)
HFC-134a (ppt)	6.1 (±6.9)	8.3 (±8.1)	7.4 (±7.1)	7.4 (±7.1)
HCFC-22 (ppt)	8.7 (±26.6)	11.1 (±10.0)	8.0 (±7.2)	10.0 (±26.6)

Main findings from experiment 1:

- The sensitivity of calculated enhanced mixing ratios to the background varies among compounds (Fig. 4 and Table 1).
- The difference in derived ΔC from various background approaches is significant, suggesting, if the choice of background was inappropriate, it may result in significant biases in regional emission estimates.

Evaluation of Background Approach (4)

Experiment 2: We chose a remote site at Estevan Point, British Columnbia (ESP) (Fig. 3) to examine whether the background derived from approach (4) well represents the true background.

Our hypothesis:

If both curtain and transport are perfect, the derived ΔC at a remote site should be close to 0 and the correlation of ΔC between two anthropogenic species should collapse.

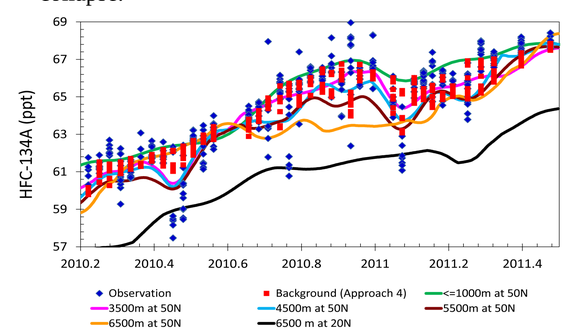


Fig. 5. Observed and calculated background mixing ratios of HFC-134a s at ESP.

Main findings from experiment 2:

- The variability of observed concentrations can not be completely explained by the background (Fig. 5).
- The correlation of ΔC between HFC-134a and HCFC-22 was improved when using approach (4) to derive background.

Experiment 3: We ran the transport model (HYSPLIT) back for 10 days and retrieved the particle locations at the end of 0, -1, -2, -3, -5, -7 and -10 days to examine in which scenario, the low mixing ratios in Fig. 5 can be best explained.

Table 2. Calculated enhanced mixing ratios (ΔC) of CO, CH_4, HCFC-22 and HFC-134a from various scenarios.

Scenarios	ΔC_{CO} (ppb)	ΔC_{CH4} (ppb)	ΔC_{HCFC22} (ppt)	$\Delta C_{HFC134a}$ (ppt)
0 day	0.4 (±11)	-1 (±12)	0.0 (±1.7)	-0.1 (±0.9)
-1 day	1.4 (±11)	2 (±14)	0.4 (±1.7)	0.1 (±1.3)
-2 day	3.0 (±11)	4 (±13)	0.7 (±1.8)	0.2 (±1.2)
-3 day	3.2 (±11)	3 (±12)	0.7 (±1.7)	0.2 (±1.1)
-5 day	4.4 (±11)	4 (±13)	1.0 (±1.8)	0.3 (±1.1)
-7 day	6.3 (±11)	8 (±13)	1.4 (±1.8)	0.5 (±1.1)
-10 day	8.5 (±12)	10 (±13)	1.8 (±1.8)	0.7 (±1.1)

Main findings from experiment 3:

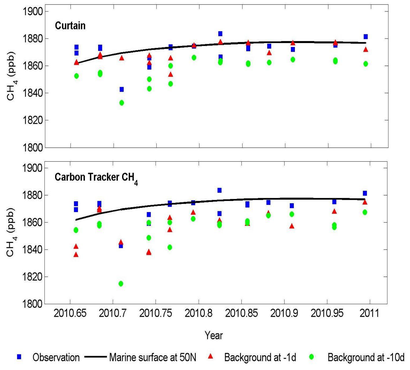
- Some of the low mixing ratios can be explained by running the model for a longer period. However, the overall variability of ΔC was not reduced (Table 2).
- The mean ΔC (the "bias") increases as we retrieve the endpoint locations further back in time (Table 2). This raised a question about whether the "curtain" can not represent background concentrations at other longitudes or the mixing ratios at the end of -10 days are no longer representative for those observed at ESP.

Experiment 4: Instead of using the empirical "curtain", we used 3D CH_4 mixing ratios derived from a global inversion model (carbon tracker – CH_4) to examine whether the "bias" observed during experiment 3 could be removed.

Table 3. Calculated enhanced mixing ratios (ΔC) of CH_4 at ESP during 2010.5 – 2011 using background derived from the "curtain" and "carbon tracker-CH_4", along with particle locations at -1, -2, -3, -5, -7 and -10 days of the air back-trajectories.

Scenarios	ΔC_{CH4} (ppb)	
	Surface Data - Using Curtain	Surface Data - Using CT-CH_4
-1 day	1.6 (±9.2)	13 (±10)
-2 day	5.1 (±8.9)	11 (±7.7)
-3 day	5.7 (±7.3)	11 (±7.7)
-5 day	7.8 (±7.2)	12 (±6.9)
-7 day	11 (±8.5)	14 (±6.6)
-10 day	15 (±5.8)	15 (±7.1)

Fig. 6. Observed and calculated background mixing ratios of CH_4 within the marine boundary layer at ESP.



Main findings from experiment 4:

- The increasing trend of the mean ΔC derived from various scenarios during experiment 3 was removed by using 3D CH_4 mixing ratios from carbon tracker (Table 3 and Fig. 6).

Conclusions

- The choice of background can influence the values of derived enhanced mixing ratios (ΔC) significantly. If ΔC was biased, it may result in a considerable bias in a regional emission estimate.
- We provided evidence that mixing ratios of a trace gas were relatively constant along an isotropic flow if there was no emission or loss during the transport. This suggests that background derived from approach (4) can well represent the true background if we have complete information about the transport and 2D background mixing ratios at the boundary of a studied region.