Inverse modeling of long-term CO emission in China with Green's function method and forward sensitivity

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1. Introduction

(a) Stationary and Mobile Sources





Emission Inventory

СТМ

Uncertainties

±70% (CO), ±31% (NOx),

±208% (BC), ±258% (OC)

INPUT

Kurokawa et al. 2013

statistics and emission factors etc.

events and time variations

(biomass burning and socioeconomic conditions etc.)

Inverse Estimate (Top-down)

• interactive (with observations and model)

Zhang et al. 2009

compensating



Spacial Resolution

Time Period

CO

CO2

regional

Heald et al., 2004 Fotems-Cheiney et al., 2011 Arellano et al., 2004

month *Yumimoto and Uno, 2006 Tanimoto et al., 2008 Wang et al., 2004*

monthly

Observation

Time Resolution)

in-situ

Yumimoto and Uno, 2006 Tanimoto et al., 2008

Forward

Wang et al., 2004 Arellano et al., 2004 Heald et al., 2004

NOx

gridded

Yumimoto and Uno, 2006 Stabrakou and Müller, 2006 Kopacz et al., 2010

Year Stabrakou and Müller, 2006 Kopacz et al., 2010 Arellano et al., 2004

daily

air craft

Wang et al., 2004

Methane

aerosols

Tanimoto et al., 2008

years Fotems-Cheiney et al., 2011

diurnal

satellite

Stabrakou and Müller, 2006 Kopacz et al., 2010 Arellano et al., 2004 Fotems-Cheiney et al., 2011

Adjoint (Backward)

Yumimoto and Uno, 2006 Tanimoto et al., 2008 Stabrakou and Müller, 2006 Kopacz et al., 2010 Fotems-Cheiney et al., 2011

Method



Forward method calculates sensitivities of sources (emissions) to receptors (measured concentrations). *We need 'ensemble' simulation for each source.



Backward method calculates sensitivities of receptors (observations) to sources (emissions).

*We need adjoint of the forward model.

In this study...





Spacial Resolution

Time Period

Time Resolution)



Arellano et al., 2004

month

monthly

in-situ

Yumimoto and Uno, 2006 Tanimoto et al., 2008 Wang et al., 2004

Yumimoto and Uno, 2006

Tanimoto et al., 2008

NOx



Yumimoto and Uno, 2006 Stabrakov and Müller, 2006 Kopacz et al., 2010

Methane

Year Stabrakov and Müller, 2006 Kopacz et al., 2010 Arellano et al., 2004

daily

air craft

Wang et al., 2004

Tanimoto et al., 2008

aerosols



diurnal



Kopacz et al., 2010 Arellano et al., 2004 Fotems-Cheiney et al., 2011

Adjoint (Backward)

Yumimoto and Uno, 2006 Tanimoto et al., 2008 Stabrakov and Müller, 2006 Kopacz et al., 2010 Fotems-Cheiney et al., 2011



Observation



Arellano et al., 2004 Heald et al., 2004

3. Method (Green's Function Method)

Menemenlis et al., 2005 Yumimoto and Uno., 2012

Observation

 $\mathbf{y}^{\mathrm{o}} = H_{\mathrm{i}}[\mathbf{x}^{\mathrm{t}}] + \boldsymbol{\varepsilon} \qquad (1)$

y^o: observation, ε: noise process

Convolution

$$\mathbf{y}^{\mathrm{o}} = G(\mathbf{\eta}) + \mathbf{\varepsilon} = G(\mathbf{0}) + \mathbf{G}\mathbf{\eta} + \mathbf{\varepsilon} \quad (3)$$

G: convolution of H and M

assuming G is linear

G: Green's function matrix, forward sensitivity

*j*th column vector of **G**:

$$\mathbf{g}_j = \frac{G(\mathbf{e}_j) - G(\mathbf{0})}{e_j}$$

miss-fit between observations and model

$$\mathbf{d} = \mathbf{y}^{\mathrm{o}} - G(\mathbf{0}) = \mathbf{G}\mathbf{\eta} + \mathbf{\varepsilon}$$

• Model

$$\mathbf{x}^{t}(t_{i+1}) = M_{i}[\mathbf{x}^{t}(t_{i}), \mathbf{\eta}]$$
(2)

x^{*t*}: true state, **\eta**: control parameter

3. Method (Green's Function Method)

Menemenlis et al., 2005, MWR

miss-fit between observations and model

 $\mathbf{d} = \mathbf{y}^{\mathrm{o}} - G(\mathbf{0}) = \mathbf{G}\mathbf{\eta} + \mathbf{\varepsilon}$ (4)

Cost function (to be minimized)

$$\mathbf{J} = \mathbf{\eta}^{\mathrm{T}} \mathbf{Q}^{-1} \mathbf{\eta} + \boldsymbol{\varepsilon}^{\mathrm{T}} \mathbf{R}^{-1} \boldsymbol{\varepsilon}$$
 (5)

Solution

$$\boldsymbol{\eta}^{a} = (\boldsymbol{Q} + \boldsymbol{G}^{T}\boldsymbol{R}^{-1}\boldsymbol{G})^{-1}\boldsymbol{G}^{T}\boldsymbol{R}^{-1}\boldsymbol{d} \qquad (6)$$

• We set η as scaling factor for emissions



Model (Chemical Transport Model; CTM)

GEOS-Chem Chemical Transport Model (CTM)



Capabilities of present standard model:

 Aerosol chemistry and microphysics, tropospheric ozone-OH-NO_x chemistry, carbon gases, mercury, hydrogen, ²²² Rn/²¹⁰Pb/⁷Be...

- 1980-present GEOS meteorological data, future and paleoclimates (GISS GCM)
- Horizontal resolution:1/2°x2/3° (native), 1°x1°, 2°x2.5°, 4°x5°
- Adjoint model for inverse/sensitivity analyses

Tagged CO simulation

calculate Green's function matrix by one integration

11 geographical tagged region

4 tags (regions) in China

emissions from the 11 geographical regions are inversely optimized

- GEOS-Chem v9-01-01
- GEOS5 Met. fields
- a priori (base) emission

FF (Fossil Fuel): EDGAR (2005) BF (Bio-Fuel) : Yevich and Logan (2003) BB (Biomass Burning): Duncan et al. (2003)

= 117.8 Tg/year from China



Observation (Constraint) MOPITT CO profile (V4)

- on board TERRA satellite
- 10:30 Local Time
- over 10-year continuous observation (2-month lack in Aug. and Sep. 2009)

200501 @ surface



200501 @ 700hPa



Monthly-averaged CO distribution from MOPITT

Observation (Constraint) MOPITT CO profile (V4)

- on board TERRA satellite
- 10:30 Local Time
- over 10-year continuous observation (2-month lack in Aug. and Sep. 2009)
- the highest sensitivity at 700 hPa



Example of averaging kernels of 1000, 900, 800, 700 and 600 hPa.

Observation (Constraint) MOPITT CO profile (V4)

- on board TERRA satellite
- 10:30 Local Time
- over 10-year continuous observation (2-month lack in Aug. and Sep. 2009)
- the highest sensitivity at 700 hPa

We use MOPITT measurements

- monthly-averaged
- measured at 4 vertical levels (surface, 900, 800, 700 hPa)
- corrected assuming 5% positive bias (Kopacz et al., 2009)
- through averaging kernel (*H* includes operation of AK)

25,000-30,000 data/month used in the inversion



MOPITT data region used in the inversion

Global 2x2.5



- 11 regional tags
- a priori = EDGAR

China Nest 0.5x0.667



• finer China tags • REAS V2

MOPITT V4 TIR Only

- Thermal Infrared Only Product
- the highest sensitivity around 700 hPa
- lower 4 vertical levels
 (Surface 900, 800, 700)
- through averaging kernel
- ~30,000 data/month used

Yumimoto and Uno, 2012

MOPITT V5 TIR/NIR

- Thermal Infrared
 - + Near Infrared Product



additional sensitivity near surface

Yumimoto et al., in prep.

vs. MOPITT (ppbv, 2D-distribution, season)



CO mixing ratio (ppbv) at 700 hPa from MOPITT, a priori, and a posteriori

A posteriori successfully re-produce CO outflows from China to Japan

vs. MOPITT (ppbv, 2D-distribution, season)

CO mixing ratio (ppbv) at 700 hPa from MOPITT, a priori, and a posteriori

vs. MOPITT (Time-Series, Statistics)

vs. in-situ measurements (independent validation)

Scatter plots of in-situ measurements vs. model

a posteriori emission (2D-distribution, season)

a posteriori emission (2D-distribution, season)

a posteriori emission (seasonal cycle)

a posteriori emission (seasonal cycle)

a posteriori emission (annual variation)

a posteriori emission (annual variation)

6. Summary

In this study,

• CO emissions in China are inversely optimized with Green's function method, MOPITT vertical profiles, and GEOS-Chem for the recent 6 years.

We found that

- a posteriori emissions successfully reproduce
- (1) CO outflows from China to East China Sea and the Japanese archipelago in winter and spring.
- (2) high CO over the central eastern China region.
- a posteriori emissions exhibit significant variation

(peak in winter-spring, bottom in summer)

• a posteriori Chinese CO sources range from 156.1 to 180.8 Tg/year, representing inter-annual variations due to socioeconomic conditions