Quantifying the sensitivity of U.S. ozone concentrations to domestic vs international emissions through coupled GEOS-Chem Adjoint and CMAQ DDM source-receptor modeling Ôr The "Boundary Sensitivity Project" Farhan Akhtar, Barron Henderson, Sergey Napelenok, Daven Henze, Susan Anenberg, John Langstaff, Rob Pinder









PAST WORK ON INTERNATIONAL TRANSPORT OF AIR POLLUTION



Sources: Fiore et. al. 2009; 2010 HTAP Report (Part A)

FUTURE EMISSION CHANGES DO NOT OCCUR IN LARGE RECTANGLES AT FIXED RATES

Spatial heterogeneity in SO₂ emissions changes ...

... following a single Representative Concentration Pathway for AR5: RCP 8.5: 2050 - 2000 ... in the difference between two Pathways for AR5: RCP 8.5 2050 — RCP 4.5 2050



High-resolution sensitivity modeling techniques may be used to evaluate inter-regional variability in emission changes.

How can global and regional highresolution sensitivity models be linked to provide information regarding the international transport of air pollution? How can global and regional highresolution sensitivity models be linked to provide information regarding the international transport of air pollution?

Global and regional modeling are linked through boundary conditions

How can global and regional highresolution sensitivity models be linked to provide information regarding the Multiyear Boundary Conditions for CMAQ 5.0 from





GEOS-CHEM EVALUATION

Comparison With Satellite Observations

Tropospheric Emission Spectrometer (TES, Beer IEEE 2006) provides a benchmark for

boundary conditions, which can be evaluated following Bowman et al. (IEEE, 2008,

In the South than the North. Average biases greater than twice the observational

are made to concentrations along the CMAQ AQMEI boundary.

Comparison To Surface Observations We evaluate GEOS-Chem modeled surface concentrations with measurement data at sites across the United States and Canada (AOS, NAPS, CSN, CASTNET and IMPROVE).

January 2006-2008

faily average values (every third day). Overall, 0, mixing ratios were within 15 pob in January, and were generally overestimated by 5-25ppb in August. Modeled SO, aeroso

oncentrations agreed closely with measurements. The largest differences occurs in August in the Northeast, For OC aerosol, modeled and measured values generally agree with the exception of rural west coast sites in January and high-elevation Northwestern sites in August, Average 2008-2008 modeled biases (modeled-observed) for each site

August 2006-200

Jammer

Inc. or it is a little

tion inset in legend). GEOS-Chem performs better in August than January and better

reartainty are indicated by colored triangles (red = high; blue=kee). These comparison

Farhan Akhtar¹, Barron Henderson², Wyat Appel¹, Sergey Napelenok¹, Bill Hutzell¹, Havala Pye¹, Kristen Foley¹ 1 - U.S. ENVIRONMENTAL PROTECTION AGENCY OFFICE OF RESEARCH AND DEVELOPMENT 2 - UNIVERSITY OF FLORIDA ENVIRONMENTAL ENGINEERING SCIENCES

ABSTRACT

sispheric transport is an important source of variability in local ant concentrations (e.g., ozone). Regional models represent where transport with boundary conditions (BC) that can be r variable. Variable BC capture trends of global emissions and instation patterns on the long-range transport, and can be derived from global simulations

This study describes an available database of hourly GEOS-Chem output (10 years) and new tool, geos2cmaq, to convert GEOS-Chem outputs (with Secondary Ontanic Aerosol extensions) into boundary and initial conditions for CMAO. The tool is compatible with SAPRCO7 and CB05 for CMAQ version 5.0.

GEOS-2-CMAO

Glo

The GEOS-2-CMAQ tool converts cond entrations from GEOS-Chem imulations with SOA extensions into boundary conditions for regional air quality modeling applications of CMAO. The flow diagram below offices all the necessary inputs and outputs for GEOS2CMAD, all of which are available from the U.S. EPA.



U.S. EPA Archived GEOS-Chem Simulations

	years
GEDS-5	2004-2010
MERIRA	2001-2008
	GEDS-5 MERIA

The archived GEOS-Chem runs, default chemical mappings and GEOS2CMAO boundary conversion tool are available for download from the U.S. FPA For more information, please contact: Farhan Akhtar - akhtar farhan@epa.gov Barron Henderson - barronh@ufl.edu Kirsten Foley - foley, kristen@epa.gov For more information on GEOS-Chem and CMAO chemical mappings, please visit:

http://wiki.seas.harvard.edu/geos chem/index.php/GEOS-Chem_to_CMAQv5.0

e would like to acknowledge the GEOS-Chen velopment teams at Harvard and Dalhousie Universities for providing technical support and data for this project.

CHEMICAL MAPPING

Boundary information is obtained from a GEDS-Chem simulation with SOA in the form of hourly-averaged trac concentrations and, optionally, species values. Species values are delumped tracers (e.g., NOx -> NO2 + NO) or short-lived compounds (e.g., radicals) that are only reliable within the troposphere. GEOS-Chem must be modified to output species information over a nested domain. Tracer values are the best source of boundary information and most of the boundary conditions come from tracer concentrations

- Factors in the mapping account for
- 1. pobC to pob unit conversion
- 2. Assignments of real compounds to mechanism species
- 3. Speciation of lumped aerosol (dust, seasait) consistent with CMAQ speciation profiles and aerosol sizes 4. Representation of species not present in GEOS-Chem based on ratios to existing compounds

Subset	of Mapping fo	r Tv	vo CMAQ Mech	anisms	
C805 gas-	GEOS-Chem Tracer		SAPRCOTT gas-	GEOS-Chem	
phase species:			phase species:	Tracer	
00	(NO + NO2 + NO2 + 2003) - (NO + NO2 + NO3 + 10023)		8	010	
048	0.5 * 1/2 * 9895		PROPINE	1/3* PRPE	
1015	0.5 * 1/4 * PRPE		TOLLENE	1/7*700	
KA.	1/7 * 10LU		NICE	MACR	
ISPO	MICH + MVK		MA	NW.	
ED4	1/2*0298		AX1	1/2*0288	
1509	1/5*ISOP		ISOPRINE.	1/5*1909	
FNR	15/3*C3H8+4/4*ALK *A2E7+4/4*MEK+1/8	+ 3/3 + 80%	AUG	1/3*0388	
Soloct I	norganic CMAQ A	ERO	to GEOS-Chem S	species:	
,	3046	0.0776 * SALC+ 0.02465 * (D672+D573+D574) + 5044			
	ecku .	8	0.89 * 304 + 0.0776 * SALA + 0.0225 * (DSF1)		
	00-42	0.99* NH4+ 0.00005*(0571)			
ANDIA		0.01 * 1014			
,	8000		0.99 * NT+0.00020 * (DST1)		
	0.00	0.01 * WIT			
	NCLK.	0.5538 * SALC + 0.01390 * (D573 + D573 + D574)			
AMOU		C DIM * SALA			
	ACA		0.0018 * SMLA+ 0.0794	0118 * SALA + 0.07940 * (2071)	
	CT IRC		0.50319*(09	0.50218*(06F1)	
AWCOK			MTa+0.0018*(0192+0593+0694)		
ASEACAIN			0.3685 * SALC		
	SOL.	0.85995*(0512+0513+0514)			
Select	organic CMAQ AL	106	to GEOS-Chem 5	pecies:	
APOCI (for	teely ACROPA)	0.898 * (0091+00P0) + 0.00075 * (0511)			
APOCI (fair	teely ACROPA)		6.005 * (0099+00P0)		
	NIC		0.999 * (8091+802PD)		
	ALC		Gats * (BOR+PORt)		
ABAZIJ			0.12* 9045		
	NC:		0.04 * 5045		
	NO.		0.32 * 5045		
,	891		0.33*(\$041+\$042)		
,	891		0.67*(\$041+\$043)		
	előe		5043		
	NCOM	0.4 *0.001 *(009-0090)			
AP	APRCONU 0.4 * 0.999 * (0091+00P0)+ 0.3043 * (051		0.0043 *(0511)		

Example Boundary Concentrations

Average boundary conditions are shown below for Ozone, 504, and iscorene Averagi is component of organi tool. Boundary conditions from the GEOSOCIMAD tool capture a high degree of spatial, vertical and





lodeled and observed 0, mbdng ratios are daytime hourly values, while SO, and OC are

0.05 * NIT
C+0.05590*(0572+0573+0574)
CLOSED * SALA
8* SMLA+ 0.07940*(06F5)
0.50318*(05F1)
L0018*(05F2+D6F3+D6F4)
0.3665 * 5440
995 * (DST2 + DST3 + DST4)
05-Chem Species:
0091+00P0)+0.00075*(0511)
0.001*(0091+00P0)
0.999 * (8091-8090)

Engineenng

ORISE

uqh

PRESENTATION OUTLINE

- 1. Primer on sensitivity modeling techniques
- 2. How forward and reverse sensitivities describe source-receptor relationships
- 3. Description of modeling of the April 2008 episode of high international transport
- 4. Case studies: Denver and New York City
- 5. Final Thoughts: How do we link global and regional sensitivity models?

CMAQ BASE MODEL



The typical application of CMAQ modeling, gridded ambient concentrations are calculated from gridded emissions using first-principle chemistry and physics.

FINITE DIFFERENCE



- Ambient concentrations are calculated after removing or perturbing emissions of a specific source.
- No changes are made to the CMAQ model though the alterations in input emissions can lead to significant changes in the chemical regime.

DIRECT, DECOUPLED METHOD



- The CMAQ model is updated to calculate the response in concentrations to a small change in emissions of a single source or group of sources.
- These sensitivities are calculated directly using similar equations as the base CMAQ model.
 - The underlying model equations and chemical regime remain unchanged.

ADJOINT MODEL



- Evaluates effect of each emission on selected concentration metric.
- Directly determines sensitivities using similar equations as the base CMAQ model.
 - Emissions and concentrations remain entirely unchanged.

RESPONSE SURFACES



Quickly indicate the effects of changing emissions (SOURCES) on pollution concentrations (RECPTORS)

RESPONSE SURFACES



DDM gives the response of all receptors to several sources in a single CMAQ run (forward sensitivities)

RESPONSE SURFACES



Adjoint models give the response of a single receptor to all emission sources at locations (reverse sensitivities)



Adjoint and reverse sensitivities are best used to understand how multiple sources impact specific receptors

DDM and forward sensitivities are best used to understand how specific sources impact multiple receptors

Adjoint and reverse sensitivities are best used to understand how *International Emissions* impact *Regional Boundaries*

DDM and forward sensitivities are best used to understand how specific sources impact multiple receptors Adjoint and reverse sensitivities are best used to understand how *International Emissions* impact *Regional Boundaries*

DDM and forward sensitivities are best used to understand how *Regional Boundaries* impact *Local Concentrations*

AQMII BOUNDARY SENSITIVITIES

APRIL 2008 EPISODE OF HIGH INTERNATIONAL TRANSPORT



- Oltmans et al. 2010 describe a period of high influence of international transport on ozone over western North America.
- Based on a GEOS-chem adjoint run of the sensitivity of US ozone concentrations to atmospheric ozone concentrations, the boundary was divided into corners representing unique source areas of ozone.
- CMAQ boundary is further divided into "upper" and "lower" regions at 0.74 sigma (bottom 20 layers, ~3km) to separate local and longdistance transport.



DENVER O₃ SENSITIVITY TO BOUNDARY O₃ CONCENTRATIONS



Simulation Hour after April 1, 2008

Ozone concentrations are most sensitive to O_3 concentrations at the Northwestern boundaries, primarily the upper boundary.

DENVER O₃ SENSITIVITY TO BOUNDARY O₃ CONCENTRATIONS



Boundary influence on Denver is dominated by the Northwest boundary

NORTHWEST O_3 SENSITIVITY TO GLOBAL NO_x EMISSIONS

Percent change in total ozone at Northwest upper boundary



NORTHWEST O_3 SENSITIVITY TO GLOBAL NO_x EMISSIONS

Percent change in total ozone at Northwest upper boundary



Percent change in total ozone at Northwest lower boundary



NORTHWEST O_3 SENSITIVITY TO GLOBAL NO_x EMISSIONS



Northwest boundaries are impacted by China, United States, Russia, and Canada.

NEW YORK O₃ SENSITIVITY TO BOUNDARY O₃ CONCENTRATIONS

New York City, NY



Simulation Hour after April 1, 2008

Ozone concentrations are mostly sensitive to O_3 concentrations at Northeastern boundaries. High period of ozone is not highly sensitive to boundary ozone.

NEW YORK O₃ SENSITIVITY TO BOUNDARY O₃ CONCENTRATIONS



Boundary influence on New York City is dominated by the Northeast boundary

NORTHEAST O_3 SENSITIVITY TO GLOBAL NO_x EMISSIONS

Percent change in total ozone at Northeast upper boundary



Percent change in total ozone at Northeast lower boundary



NORTHEAST O_3 SENSITIVITY TO GLOBAL NO_x EMISSIONS



Northwest boundaries at lower levels are impacted mostly by United States and Canada. Upper boundaries are impacted more by international emissions.

ATLANTA O₃ SENSITIVITY TO BOUNDARY O₃ CONCENTRATIONS



Boundary influence on Atlanta is highly variable, though sensitivities rarely account for a large fraction of modeled ozone.

SUMMARY

- When applied thoughtfully, HDDM and Adjoint methods can be used in concert to understand important source-receptor relationships.
- Linked global and regional sensitivity tools can be used to understand the effects of changing international sources of ozone in the United States and understand the fraction of ozone that is sensitive to inflow from the boundaries.
- The results presented here are a proof of this concept. Future work will expand the modeling period to other seasons and attempt to validate the methods with finite difference modeling.

ACKNOWLEDGEMENTS

- ORISE Postdoctoral Fellowship
- Kateryna Lapina at the University of Colorado, Boulder
- Benjamin Wells, Karen Wesson, and Bryan Hubbell at the EPA

GEOS-Chem Adjoint: International anthropogenic NO_v emission influence on upper boundaries

NW

NE



GEOS5 47L NO__an 080401 at 00:00 GMT L=1 (0.1 km)

6.25e -04 [unitless]



GEOS-Chem Adjoint: International anthropogenic NO_x emission influence on **lower boundaries**

NW

GEOS5 47L NO_x_an 080301 at 00:00 GMT L=1 (0.1 km)



NE



SW

GEOS5 47L NO__an 080401 at 00:00 GMT L=1 (0.1 km)



SE

GEOS5 47L NOv_an 080301 at 00:00 GMT L=1 (0.1 km)



GEOS-Chem Adjoint: Average sensitivity of ozone at boundary regions to international emissions, April 2008*



CMAQ-HDDM: Upper boundary influence on US ozone concentrations

Layer 1 O3_BCO3NWTc



Layer 1 O3_BCO3SWTc



Layer 1 O3_BCO3NETc



Layer 1 O3_BCO3SETc



Min = -0.000 at (11,299), Max = -0.003 at (361,28)

CMAQ-HDDM: Lower boundary influence on US ozone concentrations

Layer 1 O3_BCO3NWBc



Layer 1 O3_BCO3SWBc





Layer 1 O3_BCO3NEBc





April 1,2008 0:00:00Min= -0.000 at (11,299), Max= 0.037 at (459,5)

CMAQ-HDDM: Average sensitivity of ozone to boundary regions (ppb), April 2008*



GEOS-CHEM ADJOINT:

how International Emissions impact Regional Boundaries

CMAQ HDDM:

how Regional Boundaries impact Local Concentrations