How scientific and technical air quality modeling tools interact with policy



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Outline

- 1. Brief history of AQ policy and AQ models
- 2. Roles of AQ models in policy
- 3. AQ model output for policy applications
- 4. Source attribution and receptor modeling

The first air quality regulation?



King Edward I (1239-1307)



Monet, 1904

1273: Ordinance prohibited the use of coal in London as being prejudicial to health

1306: Royal Proclamation forbade the use of (sea) coal (Marsh, 1957)

Contemporary air quality



AQ models

Plume / Dispersion:

- describe transport and diffusion of pollutants



Boubel et al., 1994

used to estimate pollution from point sources (Sutton et al., 1932), in cities (Lettau, 1931) and even throughout the globe (Machata, 1958)

AQ models – brief history

Persistence model: today's AQ is going to be just like yesterday's

Statistical models (e.g., McCollister and Wilson, 1975):

- based on timeseries, temperature, insolation, wind...
- hard to make predictions beyond range of existing data



Fig. 2. Scatterplots of forecast vs observed daily maximum ozone concentrations for the summer of 1986 at station T9 using: (a) D/S, (b) ARIMA (1,1,0), (c) TEMPER and (d) persistence models. Solid line indicates perfect prediction while the dashed line is an ordinary least squares regression estimate.

Robeson and Steyn, 1990

Timeseries+stocha

stic

AQ models

Photochemical air quality models / chemical transport models (CTMs) developed >40 years ago (e.g., Friedlander and Seinfeld, 1969)

- 1st generation: simple chemistry at local scales
- 2nd generation: local, urban, regional addressing each scale with a separate model and often focusing on a single pollutant.
- 3rd generation: multiple pollutants simultaneously up to continental scales and incorporate feedbacks between chemical and meteorological components.
- 4th generation: extend linkages and process feedback to include air, water, land, and biota to simulate the transport and fate of chemical and nutrients throughout an ecosystem.

AQ models

Current advances in AQ models:

- online / coupled simulations of chemistry and meteorology (Baklanov et al., 2014)
- Large Eddy Simulations with chemistry (LES)
- ensembles / probabilistic
- self correcting / data assimilation (Bocquet et al., 2015)

Air quality modeling for policy assessment





Roles of AQ models in policy

Setting standards

- developing concentration-response relationships
- exposure assessment / evaluating risk
- determining background / natural pollution levels

Background O₃



 $USB = O_3$ in the absence of anthropogenic emissions from the U.S.

NAB = O_3 in the absence of anthropogenic emissions from the U.S., Canada and Mexico

Background O₃

NAB O₃ from multi-model study (Lapina et al., 2014):



Background MDA8 O₃:

- Correlated with MDA8 O3 (Zhang et al., 2011)
- Relative background amount fairly consistent across models (56-67% of total daytime O_3 across entire US).

Roles of AQ models in policy

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AQ management

- Development of control strategies (e.g., SIP)
- Permitting
- Determining exceptional events and long-range transport
- Forecasting

Evaluating impacts of emissions regulations



Figure 1. Estimated annual average PM_{2.5} concentration (µg/m³) in the YRD domain in the base-case scenario.

Zhou et al., 2011



Evaluating impacts of emissions regulations

Table 1. CMAQ simulation scenarios targeting NO_{x} and other pollutant emission reductions in different sectors.

Scenario	Sector	Pollutants reduced	Reduction
1	Power	NO _x alone (SCR alone)	85%
2	Power	$NO_x + SO_2$ (SCR + FGD)	85% for $NO_x + 90\%$ for SO_2
3	Mobile	NO _x alone	20%
4	Mobile	VOC alone	20%
5	Mobile	$NO_x + VOC + PM$	20%
6	Mobile	$NO_x + VOC + PM$	50%
7	Industry	NO _x alone	20%
8	Industry	VOC alone	20%
9	Industry	$NO_x + VOC + PM$	20%
10	Domestic	NO _x alone	20%

Abbreviations: FGD_fluidized cas desulfurization: SCR_selective catalytic reduction

Table 4. Mortality change estimates for control scenarios by sector and pollutant.

Scenario	Sector	Pollutant controlled	Emission reductions from base–case (1,000 tons/year)	PM-related mortality change per year
1	Power	NO _x	610	2,000 (200, 4,000)
2	Power	SO ₂	1,300	12,000 (1,200, 24,000)
3	Mobile	NO _x	83	260 (26, 520)
4	Mobile	VOC	200	380 (38, 750)
5	Mobile	Primary PM	6	620 (62, 1,200)
7	Industry	NO _x	96	300 (30, 610)
8	Industry	VOC	300	310 (31, 610)
9	Industry	Primary PM	110	12,000 (1,200, 24,000)
10	Domestic	NO _x	15	-21 (-2, -42)

Zhou et al., 2011



Designing emissions strategies to meet a particular attainment goal

Counties projected to exceed 1st (70 ppb) and 2nd (13 ppm-hrs) NAAQS standards in Baseline 2020



Designing emissions strategies to meet a particular attainment goal

Counties projected to exceed 1st (70 ppb) and 2nd (13 ppm-hrs) NAAQS standards in Projected 2020



Exceptional events

Criteria To Be an Exceptional Event

- The event is not reasonably controllable or preventable
- The event is caused by human activity that is unlikely to recur at that location or is a natural event
- There is a clear causal relationship between the event and the monitored concentration
- The event is associated with a measured concentration in excess of normal historical fluctuations
- There would have been no exceedance or violation but for the event

Examples of Exceptional Events

- High Wind Dust Events
- Wildfire Events
- Volcanic and Seismic Activities

Exceptional events

FIGURE 1 – COLORADO COMMUNITIES AFFECTED BY THE OCTOBER 2003 EVENT



Region most affected by Wildfire Smoke

CDPHE, 2006

Forecasting

http://airquality.weather.gov



Atmospheric Environment xxx (2012) 1-24



Review

Real-time air quality forecasting, part I: History, techniques, and current status Yang Zhang^{a,b,*}, Marc Bocquet^{c,d}, Vivien Mallet^{c,d}, Christian Seigneur^d, Alexander Baklanov^e

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Assessment

- Reanalysis
- co-benefits

Assessment of air quality regulations

Impacts of CAA, 2020 - 1990



Assessment of air quality regulations

Impacts of CAA, 2020 - 1990



Change in surface annual ave PM_{2.5}

■Benefits EPA, 2011

Costs

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AQ output for policy applications

What output do air quality models generate that policy actually uses?



Russell, 1997

AQ output for policy applications

 $[O_3]$, $[PM_{2.5}]$, [Pb], $[CO_2]$,...

AQ output for policy applications: O_3 metrics

Primary:

- MDA8 = maximum daily 8-hr average
- Jerrett09 = maximum 6 month mean of daily 1-hr max



O₃ gradients near the surface

Model "surface" of GEOS-Chem actually at ~60m. Values should be reduced at canopy height (2m).



Estimated crop losses from O_3 : differences across models and metrics



	Lapir	na et al.,	2016
5°	x 0.667°	GEOS-0	Chem
(La	apina)		

2.8° x 2.8° TOMCAT (Hollaway et al., 2012)

2.8° x 2.8° MO7ART-2 (Avnery et al., 2011)

Cross-metric differences larger than cross-model differences

Notes:

0.

- Hollaway and Avnery for NA, Lapina just for US
- Hollaway and Lapina bias corrected
- All: adjust model estimates for canopy height

AQ output for policy applications: PM_{2.5}

Many models (e.g., GEOS-Chem) don't even report PM_{2.5}

- requires assumptions about size that may not be rigorously treated
- for comparison to monitors, requires equilibration to monitor RH



yearly average surface wet PM25 concentration - yearly average surface dry PM25 concentration (ug/m3)





Estimating future concentrations

Model estimates may be not be correct.

Use models in a relative fashion to minimize systematic model bias.

EPA recommended approach:

- DV_i = Design Value upon which a standard is based at location *i* (e.g., monitored 3 yr average of annual 4th highest daily maximum 8-hr O₃)
- RRF_i = Relative Response Factor from sensitivity modeling (fractional change in simulated DV)

 DV_i (future) = DV_i (baseline) $x RRF_i$
Estimating future concentrations

CAMx simulations of O_3 W126 in 2025: 70 ppb primary



Attainment of a primary standard of 70 ppb would ensure attainment of a secondary stand of 17 ppm-hrs → secondary standard = primary standard

EPA, draft RIA, 2014

AQ output for policy applications: resolution

Previous works have investigated large-scale impacts:

- global reductions to fossil and biofuel (Jacobson, 2010)
- global BC and CH₄ measures (Anenberg et al., 2011; 2012)
- widespread adoption of vehicle standards (Shindell, 2011)

However, coarse models have trouble estimating exposure:

(a) population



(b) $2^{\circ} \times 2.5^{\circ} \mod PM_{2.5}$



AQ output for policy applications: resolution

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However, coarse models have trouble estimating $PM_{2.5}$ exposure:



Individual components of $PM_{2.5}$ underestimated by 5-40% in a 2°x2.5° simulation over the US. Impacts of resolution *not* globally heterogeneous.

AQ output for policy applications: resolution

Coarse models are better at estimating long-term O_3 exposure:



At some scale, uncertainty dominated by other factors (Thompson and Selin, 2012)

AQ models still have other difficulties related to exposure, e.g.:

- resolving response in high-NOx or low-NOx environments
- near roadway PM exposure
- indoor / outdoor exposure

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Source apportionment methods

Methods implemented in common US models:

Model	Methods			
CMAQ	DDM	HDDM	Adjoint	Tagging (TSSA)
CAMx	DDM	HDDM	Tagging (OSAT, APCA, PSAT, OPSA)	
WRF-Chem	Lagrangian (WRF-STILT)		Tagging	
GEOS-Chem	Tagging	Adjoint		
AM3	Tagging			
Acronym			Definition	
DDM	Decoupled Direct Method			
HDDM	High-order Decoupled Direct Method			
TTSA	Tagged Species Source Apportionment			
OSAT	Ozone Source Apportionment Tool			
PSAT	Particulate Source Apportionment Technology			
APCA	Anthropogenic Precursor Culpability Assessment			
OPSA	Online Particulate Source Apportionment			

Source apportionment uses for policy

http://preview.wiscwebcms.wisc.edu/aqast/source-apportionment-methods.htm



Source-apportionment methods

6-30-16 DRAFT ONLY do not cite or quote



An example of surface ozone attributed to the emissions from Maryland (left) and Ohio (right) at 2 p.m. on 7 July 2011 using CAMx v6.10.

Source apportionment uses for policy

What is the spatio-temporal distribution of pollution owing to a few broadly defined or aggregated source sectors or species, such as all transportation versus power plant emissions?

What is the origin of air pollution occurring in a particular location?

What would be the impact of implementing control strategies in a different order, or the co-benefits of implementing them together over each implemented in isolation?

What are the marginal pollution responses to changes in emission (e.g., ppb per amount emitted)?

What is the complete breakdown of pollutant sources contributing to total concentration levels?

Concerns for nonlinear systems

When biogenic VOCs react with anthropogenic NOx, who is to blame for the resulting ozone?

Consider example (Dan Goldberg) of O_3 in Maryland:





Concerns for nonlinear & discontinuous systems: discontinuities

Consider metrics:

AOT40 =
$$\sum_{i=1}^{n} [C_{O_3} - 40]_i$$
 for $C_{O_3} \ge 0.04$ ppm

"critical" value after 3 months = 3,000 ppb h (Karenlampi and Skarby, 1996)

Concerns for nonlinear & discontinuous systems: discontinuities

Consider metrics:



Concerns for nonlinear & discontinuous systems: nonlinearities

W126 =
$$\sum_{i=1}^{n} \left[\frac{C_{O_3}}{1 + 4403 \exp(-0.126 \times C_{O_3})} \right] \xrightarrow{\texttt{g}}_{0.0} \xrightarrow{\texttt{$$

Consider 3 policies, that target 3 different sources, each with equal impacts on O_3 .

What are their impacts on W126?

- Depends on the order in which you consider them

What are their contributions to W126?

- Each may have relatively small contribution alone
- sum of source contributions \neq 100%

Concerns for nonlinear & discontinuous systems: higher order sensitivity analysis (HDDM)

Contributions of NOx emissions to O₃ in Atlanta (Cohan et al., 2005)



Concerns for nonlinear & discontinuous systems: response surface modeling

150 8200 7.50 7.50 7.00

13.00 12.50 12.22 12.00 11.75 11.50 11.25 11.00

6420

Wang et al.,

2011





Forster et al., 2007



Shindell et al., 2009



Henze et al., 2012



Henze et al., 2012

Speciated, country-specific contributions to global temperature change from cookstove emissions abatement

Countries with large populations using solid fuels tend to have larger OC and GHG impacts.



(Lacey and Henze, in prep)

Conclusions

- AQ models play an integral role in many stages of policy
- Thinking in advance about how AQ model results are to by used can help you tailor model output to match the policy need, for more effective transferal of information
- Many policy applications of models
 - relative changes
 - response of changes to emissions
- Care is warranted when considering source-receptor relationships and attribution for nonlinear response metrics
- Policy needs us to consider co-emitted species from controllable sectors.



Balkanov, A., et al., Online coupled regional meteorology chemistry models in Eu- rope: current status and prospects, Atmos. Chem. Phys., 14, 317–398, doi:10.5194/acp-14-317-2014, 2014.

Bocquet, M., et al., Data assimilation in atmospheric chemistry models: current status and future prospects for coupled chemistry meteorology models, Atmos. Chem. Phys., 15, 5325–5358, doi:10.5194/acp-15-5325-2015, 2015.

Bosanquet, C.H. (1936) The Spread of Smoke and Gas from Chimmneys. Trans. Faraday Soc. 32:1249. EPA, The benefits and costs of the clean air act from 1990 – 2020, Summary Report, 2011.

EPA, O3 NAAQS Regulatory Impact Analysis, 2011.

EPA, O3 NAAQS Integrated Science Assessment, 2013.

Forster, P., et al., Constituents and in Radiative Forcing, in: Climate Change 2007: The Physical Science Basis. Contributions of working group I to the fourth Assessment Report on the Intergovernmental Panel on Climate Change.

Friedlander and Seinfeld, A dynamic model of photochemical smog, ES&T, 3(11), 1175-1181, 1969.

Lapina, K., **D. K. Henze**, J. B. Milford, M. Huang, M. Lin, A. M. Fiore, G. Carmichael, G. G. Pfister, and K. W. Bowman (2014), Assessment of source contributions to seasonal vegetative exposure to ozone in the U.S., *J. Geophys. Res.*, 119, 324-340, doi:10.1002/2013JD020905.

Lapina, K., **D. K. Henze**, J. B. Milford and K. Travis (2016), Impacts of foreign, domestic and state-level emissions on ozone-induced vegetation loss in the U.S., *Environ. Sci. Technol.*, 50 (2), 806-813, doi: 10.1021/acs.est.5b04887.

Lettau, H., Die wirksamkeit einer grosstadt als qulle von luftverschmutzung, Gerlands Beitr. Geophys., 31, 387-397, 1931.

Machata, L., Global scale dispersion of the atmosphere, Proc. U.N. Int. Conf. Peaceful Uses At. Energy, 2nd, 519-523, 1958.

Marsh, A., Air pollution legislation, Air Pollution, Ed. M. W. Thring, pp 239 – 45, 1957.

Robeson and Steyn, Evaluation and comparison of statistical forecast models for daily maximum ozone concentrations, Atmos. Environ., 24B(2), 303-312, 1990.

Russell, A., Regional photochemical air quality modeling: Model formulations, history, and state of the science, *Annu. Rev. Energy Environ.* 1997. 22:537–88.

Sutton O.G. (1932) A theory of Eddy Diffusion in the Atmosphere. Proc. Roy. Soc. A, 135:143.

Zhang, L., D. J. Jacob, N. V. Downey, D. A. Wood, D. Blewitt, C. C. Carouge, A. van Donkelaar, D. B. Jones, L. T. Murray, and Y. Wang (2011), Improved estimate of the policy-relevant background ozone in the United States using the GEOS-Chem global model with 1/2 x 2/3 horizontal resolution over North America, Atmos. Environ., 45(37), 6769 – 6776, doi:10.1016/j.atmosenv.2011.07.054.

Zhou, Y., J. S. Fu, G. Zhuang, and J. I. Levy, Risk-based prioritization among air pollution control strategies in the Yangtze River Delta, China, EHP, 118(9), 1204-1210, 2010.

Future vegetative O₃ exposure in *Western US* following changes in emissions...



Vegetative O₃ exposure in Western US following RCP emissions



RCP 2.6: Domestic emission reductions drive attainment.

RCP 8.5: Background W126 O_3 overtakes domestic by 2020, driven largely by global CH_4 emissions.

Lapina et al., 2015

Vegetative O₃ exposure in Western US following RCP emissions



RCP 2.6: Global CH_4 emissions reductions shifts attainment forward by a decade.

Lapina et al., 2015

Vegetative O₃ exposure in Western US following RCP emissions



RCP 2.6: Global CH_4 emissions reductions shifts attainment forward by a decade.

RCP 8.5: Global CH_4 emissions increases more than counteract domestic O_3 controls.

Lapina et al., 2015

Source attribution of PM_{2.5} related global mortality

PM_{2.5} subgrid variability (0.1° x 0.1°) resolved using MODIS, MISR, SeaWiFS AOD and CALIOP vertical profile (van Donkelaar et al., 2016):





Satellite-derived $PM_{2.5}$ shows good agreement with in situ total $PM_{2.5}$ and speciated $PM_{2.5}$ concentrations (Philip et al., 2014).

Source attribution of PM_{2.5} related global mortality

PM_{2.5} subgrid variability (0.1° x 0.1°) resolved using MODIS, MISR, SeaWiFS AOD and CALIOP vertical profile (van Donkelaar et al., 2016): (a) population (b) 2° x 2.5° model PM_{2.5} (c) satellite-downscaled PM_{2.5}



Combined high-resolution $PM_{2.5}$ data and adjoint modeling affords source attribution world-wide (Lee et al., 2015).



Climate and Clean Air Coalition (CCAC)





- Initiated Feb 2012
- Bangladesh, Colombia, Ghana, Mexico, Sweden, US, and UNEP
- now 109 members (50 countries, European Commission, multiple NGOs).
- US involvement through the State Department and EPA.
- SLCP Task Force Bill introduced to Congress (May 20, 2013).

Objectives

- Raising awareness of SLCP impacts and mitigation strategies
- Enhancing and developing new national and regional actions
- Promoting best practices and showcasing successful efforts
- Improving scientific understanding of SLCP impacts & mitigation strategies

Decision Support for Global Initiatives

Climate and Clean Air Coalition (UN)

 Reducing BC, CH₄ and other emissions from vehicles, brick production, oil & gas, solid waste

Cross-cutting efforts:

- Financing SLCP mitigation
- SLCP National Action Plans



www.unep.org/ccac

First action: rapid emission and scenario assessment toolkit



Use country-specific responses for arbitrary **Demissions** from adjoint calculations

Air quality modeling: chemical transport model

GEOS-Chem (Bey et al., 2001; Park et al., 2004)

Reactive convection-diffusion:

$$\frac{\partial c_i}{\partial t} + \mathbf{u}\nabla c_i = \frac{1}{\rho}\nabla(\rho K \nabla c_i) + \frac{1}{\rho}f_i(\rho \mathbf{c}) + E_i$$

 C_i = mixing ration of species *i* f_i = mass action of species *i* **U** = wind E_i = emission of species *i*

Energy systems optimization with MARKAL

MARket ALocations (Loulou et al., 2004)

- Selects the optimal mix of technologies and fuels at each time step to minimize the net present value of energy system capital and O&M costs
- Subject to:
 - Current and projected technology costs and efficiencies
 - Resource supply costs and competition for fuel across sectors
 - Resource supply constraints
 - Trade costs and constraints
 - Emission limits (e.g., policies)

EPA 9-region database



EPA Nine-Region MARKAL Database

Technology Detail: Light Duty Vehicles



US climate and AQ co-benefits

Incorporate "fees" on emissions into MARKAL

- criteria pollutant fees from Hidden Costs of Energy (NRC, 2010)
- CO2 fees from Social Cost of Carbon (SCC, 2013)

AQ and Climate fee-based **co-benefits** (Brown et al., 2013; in prep):



- Air quality co-benefits of GHG policies are more prominent.
- Have you considered the air quality impacts of your renewable energy source?

US climate and AQ co-benefits: GLIMPSE and MARKAL

AQ and RF diagnostics of emissions-cap scenarios (Akhtar et al., 2013):



AQ and Climate fee-based co-benefits (Brown et al., 2013; in prep):



Climate and health impacts of Short Lived Climate Pollutants (SLCPs)

 $SLCPs = CH_4$, **BC**, OC, CO, VOCs, NO_x, SO₂, NH₃, (HFCs)



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Integration of climate impacts into design of air quality control strategies

Abundance-based RF



Emissions-based ΔT



The traditional IPCC bar chart (abundance-based radiative forcing) has been invaluable for atmospheric scientists...

...yet disconnected from the needs of policy makers, who need to know the impacts of control strategies on co-emitted species.

Refining the bar chart: from abundance-based to emissions-based RF

- Perturbing emissions & recalculating RF:
- Sector specific contributions: Fuglestvedt et al., 2008;



Sector & regional specific contributions: Unger et al., 2008



Koch et al., 2005; Unger et al., 2010; Shindell et al., 2011; 2012; Menon and Bauer, 2012

Optimal AQ control strategy design: past



McRae and Cass (1981)

Optimal AQ control strategy design: present



Mesbah and Hakami (2011)

Optimal AQ control strategy design: future



Mesbah and Hakami (2011)

Integration of multiple damages for multiobjective optimization using adjoint modeling



Additional impacts being explored with adjoint modeling:

- reactive nitrogen deposition (Paulot et al., ES&T, 2013)
- vegetative exposure to ozone (Lapina et al., 2014)
- cloud condensation nucleii (Karydis et al., GRL, 2012)

Iteratively minimize cost function analogous to 4D-VAR: $\min_{\mathbf{x}} \mathcal{J}(\mathbf{x}) = \sum_{i} d_{i}(\mathcal{M}(\mathbf{x})) + (\mathbf{x} - \mathbf{x}_{a})^{T} \mathbf{C}_{\mathbf{x}}(\mathbf{x} - \mathbf{x}_{a})$ *i damages abatement costs*

Source attribution of PM_{2.5} related mortality

Source contributions to national mortality from $PM_{2.5}$ - total estimated to be 117,000 / yr

compare to 130,000 / yr from Fann et al. (2012)

- contributions by location / sector / species:

From fossil fuel SO_2 (25,638)

From fossil fuel NO_x (19,816)



note: preliminary analysis, complete annual average results in progress

Analysis valuable for determining health impacts of future emissions control strategies, particularly jointly addressing $PM_{2.5}$ and O_3

Environmental impacts of NH₃

Estimated N deposition from NH_{x} , Dentener et al. (2006)



Areas where color approaches dark red --> deposited levels are hazardous to ecosystem.

NH₃ emissions:

- increased by factor of 2 5 since preindustrial era.
- to double by 2050 (IPCC, Denman et al., 2007; Moss et al., 2010).
- contribute to 46 Tg gap in global N budget (Schlesinger, 2009)?







HTAP: we have multiple models capable of each



Variability across approaches depends upon response metric

Concerns for nonlinear systems: cross sensitivities by region and city area



Constraining aerosol sources with 4D-Var approach

Model (GEOS-Chem, *www.geos-chem.org*) predicts 4D distributions:



4D-Var approach: constrain emissions through inverse modeling

- uses adjoint of GEOS-Chem (Henze et al., 2007)
- assimilates observations (from satellites)
- adjusts emissions (x) at the grid-scale to minimize J:

$$\min_{\mathbf{x}} \mathcal{J}(\mathbf{x}) = \sum_{i} (M(\mathbf{x}) - \mathbf{y}_{i})^{T} \mathbf{S}_{y}^{-1} (M(\mathbf{x}) - \mathbf{y}_{i}) + (\mathbf{x} - \mathbf{x}^{a})^{T} \mathbf{S}_{x}^{-1} (\mathbf{x} - \mathbf{x}^{a})$$

model error a priori constraint

 \mathbf{x} = emissions, M = model, \mathbf{y} = observations, \mathbf{S}_{y} , \mathbf{S}_{x} = error covariances

Model sensitivity relationships

Model: estimates, c_i , and parameters, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i c_i, p_i Ideally, want model Jacobian, $\frac{\partial \mathbf{c}}{\partial \mathbf{p}} = \begin{bmatrix} \frac{\partial c_1}{\partial p_1} & \cdots & \frac{\partial c_1}{\partial p_J} \\ \vdots & \ddots & \vdots \\ \frac{\partial c_I}{\partial p_1} & \cdots & \frac{\partial c_I}{\partial p_J} \end{bmatrix}$

but it is generally much too large to calculate.

Forward sensitivity



Adjoint sensitivity



Air pollution and visibility: urban scale







Pasadena, CA

(photo courtesy of M. Kleeman)

~3 Million premature deaths per year globally (GBD, 2012)

Air pollution and visibility: regional scale



Transcontinental health impacts (HTAP, 2010) Aerosols interact with visible light.