Modeling atmospheric aerosols and synergies with observations: Opportunities, challenges, and way forward

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Introduction

- Aerosols have a number of important effects on the Earth’s environment and lives: climate, weather, geochemical cycles, and air quality.
- Aerosols are mixtures of different species with the most common ones are sulfate, nitrate, black carbon (BC), organic carbon (OC), dust, and sea salt.
- They come from different sources, such as fossil fuel combustions, agriculture or forest fires, volcanic eruptions, oceans, vegetation, deserts.
- They also have different particle sizes.
- Some of them are emitted directly to the atmosphere (“primary aerosol”) but others are formed in the atmosphere (“secondary aerosol”).
- In contrast with long-lived greenhouse gases (many years), aerosols are short lived (a few days), thus they exhibit significant regional and seasonal variations.
A model can integrate the current best knowledge into a global or regional computational framework to understand the atmospheric processes, chemical, physical, and optical properties of aerosols.

A model can explain the observed quantities with physical understanding.

Only a model will be able to project the future change with the future emission scenarios.

However, the model simulated results have to be objectively evaluated against observations in order to have credibility.
All models are wrong, but some are useful.

– George Box
Today’s talk

1 Basics of modeling of atmospheric aerosols
2 Types of observations of atmospheric aerosols and Evaluation of model with different types of observations
3 Examples of recent research topics using model and observations
4 Discussion of opportunities, challenges, and way forward
1

BASICS OF MODELING OF ATMOSPHERIC AEROSOLS
From emission to concentration to climate forcing: What does a model do

- Emission
  - Anthropogenic, natural, biomass burning
  - Individual species and precursor gases

- Mass concentration
  - Individual species
  - Dry concentrations
  - Observations at certain time and vertical location

- Optical properties
  - Total extinction or absorption
  - Wavelength dependent
  - Obs. Mostly remote sen.

- Radiative effects
  - MEE
    - Particle size
    - Mixing state
    - Refractive index

Each process has substantial uncertainties and the model approach has a wide range of sophistication/simplification/parameterization
Emission

- Emission inventories based on energy use, fuel type, emission factors, environmental regulations, etc.
- Estimated from satellite/ground based observations, such as MODIS fire counts, OMI volcanic SO$_2$
- Calculated on-line in the model based on meteorological conditions (e.g., winds, temperature, RH)
- It is difficult to directly verify if the emission is accurate
Examples of emissions used by the models

Fossil fuel emission: input from emission dataset (ACCMIP)

Biomass burning emission: input from emission dataset (GFEDv3)

Volcanic emission: input from emission dataset (Carn et al., 2015)

Dust emission: Calculated in the model = f (wind, soil moisture, etc.)

Sea salt emission: Calculated in the model = f (wind)
Aerosol mass in the atmosphere

- Model calculated atmospheric mass concentration/loading are the results of atmospheric processes in the model, including emission, chemistry, transport, dry deposition, and wet removal.
- In many cases those processes are difficult to be directly verified from measurements, especially the removal rates (≠ removed amount).
- Model calculated species concentrations can be compared directly with in-situ observations.
Aerosol optical properties

- From the aerosol mass loading \( (M_{\text{dry}}) \) to AOD:

\[
AOD = MEE \times M_{\text{dry}}, \quad \text{where} \quad MEE = \frac{3Q_{\text{ext}}}{4\pi \rho r_{\text{eff}}} \cdot \frac{M_{\text{wet}}}{M_{\text{dry}}}
\]

- \( M_{\text{dry}} \) is the result from model-simulated atmospheric processes
- MEE embodies the aerosol physical (including microphysical) and optical properties
- Since \( Q_{\text{ext}} \) varies with the wavelength of radiation, so do MEE and AOD
- AOD is the most commonly used quantity retrieved from remote sensing measurements and is frequently used for model evaluation
2

TYPES OF OBSERVATIONS AND EVALUATION OF MODEL WITH THEM
Satellite remote sensing of aerosol

- A measurement-based characterization of aerosols on a global scale can be realized only through satellite remote sensing, which is the only means of characterizing the large spatial and temporal heterogeneities of aerosol distributions.
- Satellite retrieves aerosol amount based on the amount of light that is attenuated by aerosols.
- “Passive sensor”: depending on light source from the sun (or moon, or stars).
- “Active sensor”: shooting its own light (lidar) to earth.
- Passive sensors have much wider horizontal coverage than active sensors, but they offer little information on vertical profile; active sensors measures vertical distributions, but they have very small horizontal footprint.
Global distributions of AOD from passive sensors

Figure from Chin et al., 2014
AOD and particle size information

AOD from MODIS retrieval and GOCART model simulation. Figure in Chin et al., 2007. Original was from Yoram Kaufman, 2002. Red: find mode AOD; green: coarse mode AOD. The brightness of color indicates the depth of the aerosol.
Aerosol vertical profile from active sensor CALIOP

Figure from Liu et al., 2008
Ground-based remote sensing

- **Passive technique:**
  - Sunphotometer measurements of column AOD, such as those measured by AERONET
  - Data are usually considered to be “ground truth” that are used for satellite retrieval validation and model evaluation

- **Active technique:**
  - Lidar measurement of aerosol vertical profiles
In-situ measurements

- Focused field campaigns
  - With clearly defined objectives
  - Usually involves aircrafts or ships or trucks to measure compositions, chemistry, and microphysical and optical properties of aerosols
  - Supplemented by ground-based and satellite observations

- Ground measurements networks
  - Provide stable, long-term monitoring of aerosol species
Laboratory measurements

- Measuring chemical reaction rates and products in well controlled conditions
- Measuring physical and chemical properties
- Analyzing samples collected in the fields
- Testing and calibrating instruments
Model simulations evaluated with observations: Examples from AeroCom studies

- AeroCom: aerosol comparisons between observations and models
- AeroCom is an open international initiative of scientists who are interested in understanding aerosol effects on environment with global models that are evaluated by satellite and other platform data
- AeroCom was initiated in 2003 and just had the 15th annual workshop here in Beijing last week
Comparisons of AOD with satellite and AERONET

- Models are much more diverse in aerosol composition than total AOD
- Satellite data is inadequate for constraining modeled aerosol composition

<table>
<thead>
<tr>
<th>AOD at 550 nm</th>
<th>Mean</th>
<th>Median</th>
<th>Range</th>
<th>Stddev/Mean x 100%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfate</td>
<td>0.035</td>
<td>0.034</td>
<td>0.015-0.051</td>
<td>33%</td>
</tr>
<tr>
<td>Black carbon</td>
<td>0.004</td>
<td>0.004</td>
<td>0.002-0.009</td>
<td>46%</td>
</tr>
<tr>
<td>Organic matter</td>
<td>0.018</td>
<td>0.019</td>
<td>0.006-0.030</td>
<td>36%</td>
</tr>
<tr>
<td>Dust</td>
<td>0.032</td>
<td>0.033</td>
<td>0.012-0.054</td>
<td>44%</td>
</tr>
<tr>
<td>Sea salt</td>
<td>0.033</td>
<td>0.030</td>
<td>0.02-0.067</td>
<td>42%</td>
</tr>
<tr>
<td>Total AOT at 550 nm</td>
<td>0.124</td>
<td>0.127</td>
<td>0.065-0.151</td>
<td>18%</td>
</tr>
</tbody>
</table>

Figure adapted from Kinne et al., 2006
The comparisons show that models are generally have steeper longitudinal gradient of dust decreasing from east to west over the North Atlantic, implying too fast removal of dust during transport.
Comparisons of BC vertical profile with HIPPO aircraft data

- Most models overestimate BC concentrations in the free troposphere
- Model removal of BC in the free troposphere is most likely being too slow

Figure from Samset et al., 2014
Comparisons with surface aerosol concentrations at three IMPROVE sites, 2010 monthly mean

\[ \text{SO}_4^{2-} \]

\[ \text{BC} \]

\[ \text{OA} \]
Evaluation with data reveals model strengths and weaknesses and leads to model improvements

- It is very important to evaluate the models with data from many different angles
- Thorough evaluation should lead to model improvements to establish the model credibility for its applications
- Evaluation should be objective and quantitative – using the phrase like “they agree well” sounds subjective and does not provide information on “how well” is well
- AeroCom is a very attractive platform for such evaluation with extensive archived data and tools and science expertise
3

RESEARCH TOPICS WE ARE PURSUING:

A) EAST ASIAN WINTER MONSOON AND AIR POLLUTION IN CHINA
B) VOLCANIC AND ANTHROPOGENIC AEROSOLS IN THE UTLS: SOURCES AND THE ROLE OF ASIAN SUMMER MONSOON TRANSPORT
A) East Asian winter moon and air quality in China: Particle pollution is a serious problem in East Asia, especially in winter.

- **Shenyang**: Avg > 67 μg m⁻³
- **Beijing**: Avg = 102 μg m⁻³
- **Shanghai**: Avg = 60 μg m⁻³
- **Chengdu**: Avg = 97 μg m⁻³
- **Guangzhou**: Avg = 55 μg m⁻³
Modeling approach

- Using a global model simulation and meteorological data reanalysis from 1980 to 2009 to examine the role of anthropogenic emission and meteorological conditions in controlling the particle pollution levels in winter
- Examining the relationship between a few key meteorological variables with East Asian winter monsoon index (EAWMI) and discuss the feasibility of using them to predict the pollution levels in different parts of China
- Sensitivity study on aerosol effects on meteorology and feedbacks
Model and meteorological reanalysis used in this study, 1980-2009

- **Meteorological data:**
  - NASA Modern-Era Reanalysis for Research and Applications (MERRA)

- **Aerosol simulation:**
  - GOCART model driven by the meteorological fields from MERRA reanalysis for 1980-2009, $2^\circ \times 2.5^\circ$ resolution
  - Anthropogenic and biomass burning emission: A2-ACCMIP
  - Natural sources: biogenic, volcanic, dust, sea salt
Anthropogenic sulfur missions, 1980 vs. 2009
Model experiments and derived information on anthropogenic aerosol concentration change due to change of emissions or meteorology

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Emission</th>
<th>Information</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base 1980-2009</td>
<td>All emissions from anthropogenic and natural sources</td>
<td>Change of anthropogenic aerosol due to change of both anthropogenic emission and meteorology</td>
</tr>
<tr>
<td>Natural 1980-2009</td>
<td>Only emissions from natural sources included</td>
<td>Change of anthropogenic aerosol due to change of anthropogenic emission</td>
</tr>
<tr>
<td>FixedEmi 1980-2009</td>
<td>Anthropogenic emissions fixed at 2000 level</td>
<td>Change of anthropogenic aerosol due to change of meteorology</td>
</tr>
</tbody>
</table>

Product

<table>
<thead>
<tr>
<th>Product</th>
<th>Information</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1 = Base - Natural</td>
<td>Change of anthropogenic aerosol due to change of both anthropogenic emission and meteorology</td>
</tr>
<tr>
<td>P2 = Base - FixedEmi</td>
<td>Change of anthropogenic aerosol due to change of anthropogenic emission</td>
</tr>
<tr>
<td>P3 = FixedEmi - Natural</td>
<td>Change of anthropogenic aerosol due to change of meteorology</td>
</tr>
</tbody>
</table>
Multi-decadal variations of pollution PM levels over China’s five megacities

Grey: P1 (Changes due to met & emi)
Multi-decadal variations of pollution PM levels over China’s five megacities

Difference of DJF SU+BC+OM_an (μg m⁻³) wrt 2000

Shenyang (41.80N 123.40E)

Beijing (39.93N 116.38E)

Shanghai (31.20N 121.50E)

Chengdu (30.06N 104.08E)

Guangzhou (23.13N 113.27E)

SY = Shenyang; BJ = Beijing; SH = Shanghai; CD = Chengdu; GZ = Guangzhou
Grey: P1 (Changes due to met & emi)
Blue: P3 (Changes due to met)
Multi-decadal variations of pollution PM levels over China’s five megacities

Difference of DJF SU+BC+OM_an (μg m⁻³) wrt 2000

- **Shenyang** (41.80N 123.40E)
  - April 1980 to December 2005
  - $R_{met} = 0.93$, $R_{emi} = 0.44$

- **Beijing** (39.93N 116.38E)
  - April 1980 to December 2005
  - $R_{met} = 0.66$, $R_{emi} = 0.43$

- **Shanghai** (31.20N 121.50E)
  - April 1980 to December 2005
  - $R_{met} = 0.81$, $R_{emi} = 0.45$

- **Chengdu** (30.06N 104.08E)
  - April 1980 to December 2005
  - $R_{met} = 0.65$, $R_{emi} = 0.93$

- **Guangzhou** (23.13N 113.27E)
  - April 1980 to December 2005
  - $R_{met} = 0.16$, $R_{emi} = 0.85$

Legend:
- **Grey**: P1 (Changes due to met & emi)
- **Blue**: P3 (Changes due to met)
- **Red**: P2 (Changes due to emi)
East Asian Winter Monsoon Index (EAWMI)

- There are several “typical” EAWMI calculated from SLP, geopotential height, winds at chosen altitudes, etc.
- We use the Jhun and Lee (2004) index in this work, which is the difference of zonal wind speed at 300 hPa between [27.5-37.5°N, 110-170°E] and [50-60°N, 80-140°E]
- Note that all EAWM indices can only well represent the monsoon characteristics over part of the East Asia, because the large domain of East Asia with complex and different climate zones

Relationships among pollution PM, PBLH, winds, and EAWMI: model results with fixed anthropogenic emission (meteorology-induced changes of pollution PM), winter 1980-2009

- PBLH and near-surface winds (indicated by W10m) are the most influential meteorological variables affecting pollution PM concentrations
- Both PBLH and W10m are positively correlated to EAWMI
- EAWMI could be used to estimate the pollution PM level change
Aerosol effects on meteorology through aerosol-radiation interaction

- Simulation of winter 2010 using the GEOS-5 AGCM with GOCART aerosol grid-components that interact with radiation (i.e., the so-called semi-direct effects)
- Driven by prescribed sea surface temperature
GEOS GCM simulations: Changes due to aerosol-radiation interaction (ARI)

Concentration

PBLH

W10m

Change due to ARI
Summary for A)

- **Wintertime pollution in megacities – emission or meteorology?**
  - Shenyang: Meteorology is most responsible for the change of pollution PM level
  - Beijing and Shanghai: Both meteorology and emissions are responsible
  - Guangzhou and Chengdu: anthropogenic emission is mostly responsible

- **What are the most important meteorological variables affecting pollution levels?**
  - PBLH and near surface winds (using W10m as an indicator)
  - Both PBLH and W10m are positively correlated with the EAWMI in eastern China
  - => Weaker EAWM associated with shallower PBLH, lower wind speed, and higher pollution PM in eastern China

- **What is the effects of pollution on meteorology that may cause further worsening of air quality?**
  - AGCM experiment has shown a “positive feedback loop” – through ARI, absorbing aerosol causes shallower PBLH and weaker winds to trap more pollutants in pollution regions at the surface
B) Natural and anthropogenic aerosols in the UTLS: Sources and the role of Asian summer monsoon transport

- The origin and variability of stratospheric aerosol have drawn considerable attention because the change of such aerosol could have long-term climate effects.

- Recent observations seem to suggest that the stratospheric aerosol has been increasing in the past decade without major volcanic eruptions.
  - Is the increase due to the Asian anthropogenic emission?
  - Or volcanoes?
Anthropogenic emission

- Anthropogenic SO$_2$ (and other pollutants as well) emissions in East Asia and South Asia have increased significantly in the last decade.
- EAS emission is much higher than SAS.
- The question is: How efficient the transport is to lift surface pollution to the UTLS?

(Figures from Chin et al., 2014)
Volcanic emissions release SO$_2$ usually at higher altitudes than anthropogenic emissions to have a more direct influence in the UTLS.
Model simulations

- Model simulations:
  - GEOS-5 AGCM, 2.5°lon x 2° lat horizontal resolution, 72 vertical layers
  - Anthropogenic and biomass burning emission: ACCMIP (Granier et al., 2011)
  - Volcanic emission: OMI-based sporadically erupting volcanic emission (Carn et al., 2015) + continuously erupting volcanic emission (Andres and Kasgnoc, 1998)
  - Sulfate from OCS oxidation included
  - Aerosols identified from (1) sporadically erupting volcanic source, (2) stratospheric background source (OCS oxidation), and (3) other (anthropogenic + biomass burning + non-volcanic natural + continuously erupting volcanic sources)

- Time period of this study: 2003-2014
Comparison with satellite aerosol data

- **OSIRIS:**
  - V5-07 level-3 monthly zonal averages at 5° latitude resolution and 1-km vertical resolution from 0-40 km (provided by U. Saskatchewan group, POC: Landon Rieger)
  - Merged SAGE-II and OSIRIS: extinction at 525 nm

- **CALIOP:**
  - Stratospheric aerosol V6, monthly zonal average extinction at 532 nm at 5° latitude resolution, 8 to 40 km (provided by Jean-Paul Vernier, LaRC)

- **SCIAMACHY:**
  - V1.1. level 3 monthly averages at 5°x5° horizontal resolution and 1-km vertical resolution from 9-40 km (provided by U. Bremen group, POC: Alexei Pozanov)
  - 550 nm extinction was interpolated from 470 and 750 nm using the Angstrom Exponent
Zonal mean aerosol extinction at 550 nm (Mm$^{-1}$), 20N-40N

**H43F2000gocart2 zonal mean aerosol extinction (550 nm) (Mm$^{-1}$) 20N-40N**

- **All aerosol**
- **Model**

**SAGE-II_OSIRIS merged zonal mean aerosol extinction, 525 nm (Mm$^{-1}$), 20N-40N**

- **OSIRIS**

**CALIOP zonal mean aerosol extinction, 532 nm (Mm$^{-1}$), 20N-40N**

- **CALIOP**

**SCIAMACHY zonal mean aerosol extinction, 550 nm (Mm$^{-1}$), 20N-40N**

- **SCIAMACHY**

### Volcanic Eruptions

1. Manam
2. Rabaul
3. Okmok/Kasatochi
4. Sarychev
5. Merapi
6. Nabro
7. Kleut

**Model All**

**Model Sporadical Volc**

**Model TropSrc + Continuous Volc**

**Model OCS_oxid**
Zonal mean aerosol extinction at 550 nm (Mm$^{-1}$) 0-20N

- **Model All**
- **OSIRIS**
- **CALIOP**
- **SCIAMACHY**
- **Sporadical Volc**
- **TropSrc + Continuous Volc**
- **OCS_oxid**

1. Manam
2. Rabaul
3. Okmok/Kasatochi
4. Sarychev
5. Merapi
6. Nabro
7. Kleut

Zonal mean aerosol extinction at 550 nm (Mm$^{-1}$), 0-20N
Source attribution – volcanic, anthropogenic, and background

- Overall, the volcanic aerosol dominates the stratospheric aerosol loading even without Pinatubo-scale large eruption (4-5 higher than anthropogenic)
- Near the tropopause, anthropogenic aerosol shows a well organized, repetitive seasonal cycle
- The “background” sulfate aerosol from OCS oxidation is the most important aerosol source >25 km
Source attribution – volcanic, anthropogenic, and background

- Overall, the volcanic aerosol dominates the stratospheric aerosol loading even without Pinatubo-scale large eruption (4-5 higher than anthropogenic)
- Near the tropopause, anthropogenic aerosol shows a well organized, repetitive seasonal cycle
- The “background” sulfate aerosol from OCS oxidation is the most important aerosol source >25 km
Seasonal variations of deep convective transport: Average OLR and UT aerosol anomaly, 5-30N

- In the subtropical northern hemisphere, the most pronounced convective features are the Asian summer monsoon and the North American summer monsoon.
- The monsoon convections pumps aerosols to the upper troposphere, even though the convection always associated with heavy rainfall.
- Composition of UT aerosol varies from year to year depending on the variation of aerosol sources.
Maximum aerosol in ATAL

Vernier et al., 2015
By model experiments separating anthropogenic and natural sources, we have found that
- volcanic aerosol dominates the total stratospheric aerosol amount even without very large volcanic eruptions like Pinatubo
- anthropogenic aerosol exhibits well organized seasonal cycle in the tropopause region

Strong summer monsoon convection in the subtropical northern hemisphere making transport of aerosols to UTLS most effective in the summer
- SAS anthropogenic aerosol dominates the ATAL aerosol in summer (60-70%), even though EAS anthropogenic emission is much higher than SAS
- Not all aerosols in ATAL are from Asia – other regions contribute as well
4

OPPORTUNITIES, CHALLENGES, AND WAY FORWARD
Opportunities

- Aerosol has a wide range of effects, from pollution to climate change, and many of those effects are still not well understood.
- There have been unprecedented rich atmospheric observations of aerosols from satellite, ground-based, and in-situ observations that are publically available to provide unlimited research opportunities.
- There have also been number of global and regional models publically available to the community to use for aerosol research.
Challenges

- No data or models are perfect – many times they are difficult to manipulate or understand.
- Satellite data have large spatial and temporal coverage but they are limited in retrievable physical quantities.
- In-situ data are more detailed in aerosol characteristics but they are limited in spatial or temporal coverage.
- Models are getting more and more sophisticated, but many processes are not observable to be evaluated with observations. They can also be computationally demanding.
Way forward

- Take ynergistic approach between modelers and observationalists, understand how to use the data and model
- Work with your colleagues and reach out to and be involved in the larger community (good example: AeroCom and AeroSat)
- Think globally, act locally – Build a view of big pictures but focus on solving one problem at a time