ADJOINT INVERSION OF ATMOSPHERIC DUST SOURCES FROM SATELLITE OBSERVATIONS

by

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FROM SATELLITE OBSERVATIONS

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Abstract to be filled ...

DEDICATION

To the memory of my grandparents

Zhaoxiang Xu

and

Shi Zhao Xu

ACKNOWLEDGMENTS

Arma virumque cano, Troiae qui primus ab oris Italiam, fato profugus, Laviniaque venit litora, multum ille et terris iactatus et alto vi superum saevae memorem Iunonis ob iram; multa quoque et bello passus, dum conderet urbem, inferretque deos Latio, genus unde Latinum, Albanique patres, atque altae moenia Romae.

GRANT INFORMATION

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CHAPTER 1

INTRODUCTION

1.1 Background and Motivation

Mineral dust represents the second productive component of atmospheric aerosol following after the sea-salt aerosol [*Textor et al.*, 2006]. Naturally, these mineral particles are produced mainly by the aerolian wind ersion in arid and semiarid areas. Anthropogenic sources of mineral aerosols include road dust and mineral dust due to land changes by human activities [References check with my proposal]. Mineral aerosols can place important impacts on the Earth system through interations with atmospheric chemistry, solar and terrestrial radiaton, clouds, and biosphere [*Shao et al.*, 2011]. An accurate representation of dust cycle in the Earth climate model is thus critical to assess these impacts. However, significant uncertainties prevail in quantifying mineral dust sources due to poor understanding of dust uplifting mechanisms and the lack of in situ measurements over the desert region.

Parameterization of processes such as saltation bombardment and sandblasting in a chemistry transport model (CTM) requires knowledge of many parameters that are poorly characterized, including surface wind speed, soil moisture, soil texture, and surface state [*Tegen and Fung*, 1994; *Ginoux et al.*, 2001; *Zender et al.*, 2003]. Not surprisingly, recent estimates in CTMs span from a few hundreds to over 4000 Tg for annual global dust emissions [*Huneeus et al.*, 2011] and can vary by a factor as large as 10 at regional scales for the same dust event(s) [*Uno et al.*, 2006]. An observation-based approach, therefore, is needed to reduce these large uncertainties in estimate of dust emissions and further improve the global modeling of atmospheric dust distribution and their impacts.

1.1.1 Impacts of dust aerosols

Atmospheric aerosols play a crucial role in the global climate change. They affect earth energy budget directly by scattering and absorbing solar and terrestrial radiation, and indirectly through altering the cloud formation, lifetime, and radiative properties [Haywood and Boucher, 2000; Ramanathan et al., 2001]. However, quantification of these effects in the current climate models is fraught with uncertainties. The global average of aerosol effective radiative forcing (ERF) were estimated to range from -0.1 to -1.9 Wm2 with the best estimate of -0.9 Wm2 [Boucher et al., 2013], indicating that the cooling effects of aerosol might counteract the warming effects of 1.820.19 Wm2 caused by the increase of carbon dioxide since the industrial revolution [Myhre et al., 2013]. The climate effects of aerosol particles depend on their geographical distribution, optical properties, and efficiency as cloud condensation nuclei (CCN). Key quantities pertain to the aerosol optical and cloud-forming properties include particle size distribution (PSD), chemical composition, mixing state, and morphology [Boucher et al., 2013]. While the daily aerosol optical depth (AOD) can be well measured from current satellite and ground-based remote sensing instrumentations [e.g., Holben et al., 1998; Kaufman et al., 2002], the accurate quantification of aerosol ERF is in no small part hindered by our limited knowledge about the aerosol PSD and refractive index (describing chemical composition and mixing state). To fully understand the role of aerosol particles in the global climate change, further development in observations along with retrieval algorithms for these aerosol microphysical properties from different platforms are thus highly needed [Mishchenko et al., 2004], and the focus of this two-part series study is the characterization of aerosol properties from ground-based passive remote sensing

[*Henze et al.*, 2007].

1.1.2 Parameterizations of dust emissions

1.1.3 Observations of dust aerosols

In the last decade, the in situ and satellite remote sensing observations have greatly enhanced our understanding of the spatiotemporal variations of dust aerosols.

1.1.4 Recent inverse modeling studies for improving dust emission

In parallel with the advancement of in situ and remote sensing observations of dust aerosols, techniques have been developed to use these observations as constraints on dust sources.

Koven and Fung [2008] have investigated ...

Source function can be estimated based topography [*Ginoux et al.*, 2001],

1.2 Main Goals of This Work

Based on the preceding discussions, this work aims at improved estimates of global dust emissions through adjoint integration of AOD retrievals from multiple satellite platforms (MODIS and MISR) with a CTM (GEOS-Chem). The overall goal is to conduct the satellite-based global model estimates of atmospheric dust distribution, and thereby advance the understanding of the impacts of atmospheric mineral dust on climate change and air quality. To accomplish this goal, this work pursues the following specific objectives:

- Develop a top-down numerical inversion scheme for constraining global dust emissions with a combined use of multi-platform AOD products and CTM adjoint, which also includes the sensitivity and error budget analysis for the optimization.

- Apply the inversion scheme developed in step 1 for a one year (i.e. 2008) of dust emissions with level 3 quality-controlled MODIS DB and MISR AOD products.

- A long-term (from 2001 to 2010) analysis of dust emissions will follow, along with studies on the seasonal and inter-annual variability of dust emissions, loadings, and direct radiative effects.

- Wherever possible, ground-based and field data will be used to validate and analyze the uncertainties of the inversion results.

Although the adjoint optimization technique we use is similar to that in Dubovik et al. [2008] and Yumimoto et al. [2007], this study differs from the those previous studies in that: (a) Multi-platform AOD products utilized to optimize dust emissions can provide tremendous dust information in fine spatial and temporal scales; (b) This study uses the satellite AOD retrievals only over and near dust source regions where dust has been transported a short distance with minimal influence of precipitation and anthropogenic aerosols; (c) Optimization of the long-term dust emissions is conducted for every grid box as a function of time (e.g., on the weekly or month scale). Although the criteria for separation of a natural and anthropogenic dust source is not clear and sometime controversial in the literature [Denman et al., 2007], especially when considering the climatic feedback on dust emissions [Zhang et al., 2002], we believe that satellite-based optimization of global dust emissions in the last decade could improve our modeling of dust radiative forcing and potentially illuminate anthropogenic components of dust sources and loadings, currently estimated at 0-20% though values as large as 50% has been postulated [Ginoux et al., 2011; Tegen et al., 1996; 2004; Mahowald et al., 2004,].

1.3 Organization of This Dissertation

We describe the GEOS-Chem simulation of mineral dust in Chapter 2 with emphersizing the physical parameterization of dust sources, after which we present the implements for the AOD observation operator and the adjoint capacity of dust emission within the GEOS-Chem ajoint model in Chapter 3. In chapter 4, we present a case study on optimizing the dust emission estimates from the satellite (MODIS) radiances over the eastern Asia, in which we also attempt to simutaneously constrain the anthropogenic emissions of the SO_2, NO_2, NH_3, and carboneous aerosols tegether with the dust aerosols. In chapter 5, we optimize the dust source parameterization from multi-satellite AOD products, particularly in improving the the estimates of soil erodibility and wind friction threshold for sand saltation over the northern Africa. Finally, we summarize the dissertation and outlook future work in Chapter 6.

CHAPTER 2

MODELING OF ATMOSPHERIC DUST

Overview This chapter presents how the physical process that mineral dust involved are quantitatively respresented in a chemistry transport model, i.e., the GEOS-Chem model. These processes include the uplifting of dust from soil surface, the transport within the atmosphere, and despoistion of dust to the surface.

2.1 Modeling of Dust Emission

2.1.1 Physical parameterization of the dust emission

The dust emission, aerolian wind erosion that results in production of mineral aerosols from soil grains, involves complex and nonlinear processes that are governed by the meteorology as well as by the state and properties of the land surfaces. Laboratory [Iversen and White, 1982] and field [Shao et al., 1996; Zender et al., 2003] wind tunnel studies suggested that dust is primiarily injected into the atmosphere during the sandblasting caused by the saltation bombardment [Alfaro and Gomes, 2001; Grini et al., 2002]. The clay- and silt-sized soil particles have strong inter-cohesive force... The saltation of sand-sized particles ... requires least threshold of wind speed...

The most important factors include wind friction velocity and its threshold for saltation, vegetation cover, soil minerology, and surface soil moisture.

In this study, the physical parameterization of dust emission is taken from a Dust Entrainment and Deposition (DEAD) model developed by Zender et al [2003a]. The DEAD scheme calculates the wind friction threshold (u_{*t}) as a function of the Reynolds number following Iversen and White [1982] and Marticorena and Bergametti [1995]. Three processes are also considered to modify the u_{*t}: the drag partitioning owing to the momentum captured by nonerodible roughness elements, the Owen effect, and moisture inhition. The horizontal saltation flux (Q_s) that is defined as the vertical integral of the stream-wise soil flux density is calculated following the theory of White [1979]:

$$Q_s(u_*, u_{*t}) = \frac{c_s \rho}{g} u_*^3 \left(1 - \frac{u_{*t}}{u_*}\right) \left(1 + \frac{u_{*t}}{u_*}\right)^2, \tag{2.1}$$

where, $c_s = 2.61$, ρ is the air density at surface level, and $u*$ is the wind friction velocity. Thus, it assumes the saltaion flux is quasi-lienarly the u_*^3 when u_* exceeds the u_{*t}. It also neglect the dependence of total Q_s on the soil size.

the total vertical mass flux of dust into transport bin j is

$$E_{d,j} = \begin{cases} T_0 A_m S \alpha Q_s \sum_{i=1}^{3} M_{i,j} & \text{if } u_* \geq u_{*t}, \\ 0 & \text{if } u_* < u_{*t}, \end{cases} \tag{2.2}$$

where, T_0 is a tuning factor chosen to adjust the global amount, A_m is the fraction of bare solil exposed in a model grid cell, S is called "erodiblity" or "perferential source function", α is the sandblasting mass efficiency factor which depends on the mass fraction of clay particles in the parent soil, and $M_{i,j}$ indicates the mass fraction of ith source mode carried into the jth transport mode.

2.1.2 Development of the wind speed distribution

In order to incorporate the variability of wind speed due to the subgrid scale circulations, we introduce a probability density function (PDF) of the wind speed within each grid box. The dust emission is computed according to the fraction of the PDF that exceeds the threshold value:

$$E_d = \int_{u_{*t}}^{\infty} E(u_*) p(u_*) du_*. \tag{2.3}$$

Where $E(u_*)$ is the emission as a function of the surface wind friction velocity, and $p(u_*)$ is the PDF of u_* within the grid box.

The PDF for surface wind speeds can be represented by a Weibull distribution [Justus et al., 1978] and has been used in recent studies [e.g., Grini and Zender, 2004; Grini et al., 2005; Ridley et al., 2013] to charaterize the subgrid dust emissions. The PDF of a Weibull random variable x is described by a shape factor k and a scale factor c:

$$p(x;c,k) = \frac{k}{c}(x/c)^{k-1} \exp\left[-(x/c)^k\right], \text{ for } x > 0. \tag{2.4}$$

One of the advantages in using the Weibull PDF is that it is analytically integrable with the cumulative distribution function:

$$P(x \le x_1; c,k) = 1 - \exp\left[-(x/c)^k\right]. \tag{2.5}$$

Based on above cumulative function, we cut off wind speeds with a minimum and a maximum wind speed to retain the central 98% of the wind PDF. As a result, the lower and upper limits of wind speed are respectively:

$$x_l = c\left[-\ln 0.99\right]^{\frac{1}{k}} \tag{2.6}$$

$$x_u = c\left[-\ln 0.01\right]^{\frac{1}{k}} \tag{2.7}$$

Parameters k and c can be estimated from the statistical mean $\bar{x}$ and variance σ^2 (of x), since they are related to $\bar{x}$ and σ^2:

$$\bar{x} = c\Gamma(1+1/k) \tag{2.8}$$

$$\sigma^2 = c^2\left[\Gamma(1+2/k) - \Gamma^2(1+1/k)\right] \tag{2.9}$$

Where $\Gamma()$ is a gamma function. According to Justus et al. [1978], k and c can be best estimated by:

$$k = (\sigma/\bar{x})^{-1.086} \tag{2.10}$$

$$c = \bar{x}\left[\Gamma(1+1/k)\right]^{-1} \tag{2.11}$$

Thus, the only parameter that must be supplied beyound the mean wind speed is the variance (σ^2) of subgrid wind speeds within the grid box. Cakmur et al [2004] calculated the σ^2 by incorporating information from the parameterizations of the planetary boundary layer along with dry and moist convection. Here, we follow Grini and Zender [2004] and Grini et al [2005] that assumed an approximation of k based on Justus et al. [1978]:

$$k = 0.94u_*^{\frac{1}{2}} \tag{2.12}$$

Finally, the dust emission flux is calculated by

$$E_{d,j} = A_m S \alpha \left(\sum_{i=1}^{3} M_{i,j} \right) \frac{c_s \rho}{g} \int_{u_{*t}}^{u_{*u}} u_*^b \left(1 - \frac{u_{*t}}{u_*} \right) \left(1 + \frac{u_{*t}}{u_*} \right)^2 p(u_*) du_*. \tag{2.13}$$

Where u_{*u} is the upper limit of wind speed determined by equation (2.7).

2.2 Modeling of Dust Transport and Deposition

CHAPTER 3

ADJOINT INVERSION OF DUST EMISSIONS

Overview

3.1 The Adjoint Inversion Framework

The adjoint of the GEOS-Chem model was developed specifically for inverse modeling of aerosol (or their precursors) and gas emissions [*Henze et al.*, 2007, 2009], and it is continuously improved and maintained by the GEOS-Chem Adjoint and Data Assimilation Working Group and its users (`http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_Adjoint`). The strength of the adjoint model is its ability to efficiently calculate model sensitivities with respect to large sets of model parameters, such as aerosol emissions at each grid box. These sensitivities can serve as the gradients needed for inverse modeling of aerosol emissions. Recent studies have used the GEOS-Chem adjoint with satellite observations to constrain sources of species such as CO [*Kopacz et al.*, 2009, 2010; *Jiang et al.*, 2011], CH_4 [*Wecht et al.*, 2012], and O_3 [*Parrington et al.*, 2012] to diagnose source regions for long-range transport [*Henze et al.*, 2009; *Kopacz et al.*, 2011], and to provide guidance on future geostationary observations of surface air quality [*Zoogman et al.*, 2011].

3.1.1 Inversion strategy

Let $\mathbf{x}$ denote a state vector of n parameters to be constrained and $\mathbf{y}$ an observation vector assembled by m measurements, and let $\mathbf{F}$ indicate a forward model that describes the physics of the measurement process. Then, we can express the relationship between the observation vector and the state vector as

$$\mathbf{y} = \mathbf{F}(\mathbf{x}) + \boldsymbol{\epsilon}, \tag{3.1}$$

where $\boldsymbol{\epsilon}$ is an experimental error term that includes observation noise and forward modeling uncertainty.

For this work, the observation vector $\mathbf{y}$ comprises measurements of aerosol loading, such as mass concentrations or optical depth, at any temporal and spatial scale. The components of the sate vector $\mathbf{x}$ could vary according to our inversion focus. For the inversion of aerosol emission estimats (as in the Chapter 4), $\mathbf{x}$ comprises the emission fluxes (or their scaling factors) of defined aerosol species within each grid cell of specified temporal resolution. In constrast, $\mathbf{x}$ consists of dust emitting parameters (or their scaling factors) when we tend to constrain the dust emission parameterization (as in the Chapter 5). The forward model $\mathbf{F}$ represents the GEOS-Chem that maps parameters from the state space to the observation space. The inversion of the state vector from these measurements is often an ill-posed problem due to non-linearity and limited sensitivity of these observed quantities to the constrained parameters. We need to combine additional constraints to make the problem amenable to inversion.

A propri information describes our knowledge of the state vector before measurements are applied. *A propri* constraint is commonly used to achieve a well-defined stable and physically reasonable solution to an ill-posed problem. Usually, *a propri* knowledge comprises both a mean state $\mathbf{x}_\mathrm{a}$ and its error $\boldsymbol{\epsilon}_\mathrm{a}$:

$$\mathbf{x} = \mathbf{x}_\mathrm{a} + \boldsymbol{\epsilon}_\mathrm{a} \tag{3.2}$$

Under assumption of Gaussian-distributed errors, the Maximum A Posteriori solution of equations (3.1) and (3.2) according to the Bayesian approach corresponds to the state vector that minimizes the quadratic cost function [*Rodgers*, 2000]:

$$J(\mathbf{x}) = \frac{1}{2}\left[\mathbf{F}(\mathbf{x}) - \mathbf{y}\right]^T \mathbf{S}_\mathrm{y}^{-1}\left[\mathbf{F}(\mathbf{x}) - \mathbf{y}\right] + \frac{1}{2}\gamma\left(\mathbf{x} - \mathbf{x}_\mathrm{a}\right)\mathbf{S}_\mathrm{a}^{-1}\left(\mathbf{x} - \mathbf{x}_\mathrm{a}\right), \tag{3.3}$$

where T indicates the transpose operation, $\mathbf{S}_\mathrm{y}$ is the error covariance matrix of measurements, $\mathbf{S}_\mathrm{a}$ is the error covariance matrix of the *a priori*, and γ is the regularization paramter. These two terms on the right side of equation (3.3) respresent the total sqaured fitting error incurred owing to departures of model predictions from the observations and the penalty error incurred owing to depatures of the estimates from the *a priori*, respectively. Thus, the minimization of $J(\mathbf{x})$ achieves the objectives of improving the agreement between the model and the measurements while ensuring that the solution remains within a reasonable range and degree of smoothness.

The regularization parameter γ in the calculation of $J(\mathbf{x})$ acts weights to balance the fitting error and the penalty error. Clearly, a good assignment of γ is of crucial importance for the statistically optimal solution. High values of γ can lead to over-smoothing of the solution with little improvement to the fitting residuals, while low values minimize the error term at the cost of greatly increasing the penalty term. Optimal values of γ can be identified at the corner of the so-called L-curve [*Hansen*, 1998].

In principle, solving this inverse problem is tantamount to a pure mathematical minimization procedure. The minimization of $J(\mathbf{x})$ is performed with an iterative quasi-Newton optimization approach using the L-BFGS-B algorithm [Byrd et al., 1995; Zhu et al., 1994], which offers bounded minimization to ensure the solution stays within a physically reasonable range. The L-BFGS-B algorithm requires knowledge of $\mathbf{x}$ and $J(\mathbf{x})$, as well as the gradient of $J(\mathbf{x})$ with respect to $\mathbf{x}$, $\Delta_{\mathbf{x}}J$. By linearizing the forward model F(x), we can determine $\Delta_{\mathbf{x}}J$ by

$$\Delta_{\mathbf{x}}J = \mathbf{K}^T \mathbf{S}_{\mathrm{y}}^{-1} \left[\mathbf{F}(\mathbf{x}) - \mathbf{y}\right] + \gamma \mathbf{S}_{\mathrm{a}}^{-1} \left(\mathbf{x} - \mathbf{x}_{\mathrm{a}}\right), \tag{3.4}$$

where $\mathbf{K}$ is the Jacobian matrix of $\mathbf{F}(\mathbf{x})$ with respect to $\mathbf{x}$, which is computed analytically by adjoint method in the GEOS-Chem adjoint. At each iteration, improved estimates of the state vector are implemented and the forward simulation is recalculated. The convergence criterion to determine the optimal solution is the smallness of the $J(\mathbf{x})$ reduction and the norm of $\Delta_{\mathbf{x}}J$. The iteration stops when the reduction of $J(\mathbf{x})$ is less than 1% within five continuous iterations. Then, the optimal solutions are identified corresponding to the smallest norm of $\Delta_{\mathbf{x}}J$ among these five last iterations.

3.1.2 GEOS-Chem adjoint modeling

The GES-Chem adjoint model was specifically developed for ... [*Henze et al.*, 2007, 2009]. It has been widely used to ... [references...].

It caluclates the adjoint, or the transpose of Jacobian matrix of receptor with respect to the state vector, following the ...

Based on the infrastructure of GEOS-Chem, we need to develope (1) an observation operator that maps the aerosol concentration into the observation space, (2) the capacity of calculating the adjoint with respect to dust emission flux, and (3) the capacity of calculating the adjoint with respect to parameters in dust emission scheme.

3.2 Implements of AOD observation operator

Two types of observation operator

3.3 Implements of Adjoint for Dust Emissions

3.4 Implements of Adjoint for Dust Flux Parameterization

In simple, The dust emission flux considering subgrid wind speeds in equation (2.13) can be writen

$$E_{d,j} = C_j S' \int_{u_{*t}}^{u_{*u}} Q_s(u_*, u_{*t}, b) p(u_*) du_*, \tag{3.5}$$

where $S' = S\alpha$, and $C_j = A_m \sum_{i=1}^{3} M_{i,j}$. We combine the erodibility S and sandblasting factor α, because both of them not only are related to the soil texture but also describe the strength efficiency of dust emission. Given the the state of land surface and the properties of surface soil, the dust emission is a function of S', b, and u_{*t}.

Here we implement the adjoint calculation for three parameters, i.e., S', b, and u_{*t}. This implementation reqiures the partial derivatives of $E_{d,j}$ with respect to these parameters (when $u_* \geq u_{*t}$):

$$\frac{\partial E_{d,j}}{\partial S'} = \frac{E_{d,j}}{S'}, \tag{3.6}$$

$$\frac{\partial E_{d,j}}{\partial b} = C_j S' \int_{u_{*t}}^{u_{*u}} \frac{\partial Q_s}{\partial b} p(u_*) du_*, \tag{3.7}$$

$$\frac{\partial E_{d,j}}{\partial u_{*t}} = C_j S' \int_{u_{*t}}^{u_{*u}} \frac{\partial Q_s}{\partial u_{*t}} p(u_*) du_*. \tag{3.8}$$

These gradients of Q_s in equations (3.7 and 3.8) can be calculated by

$$\frac{\partial Q_s}{\partial b} = Q_s(u_*, u_{*t}, b) \ln u_* \tag{3.9}$$

$$\frac{\partial Q_s}{\partial u_{*t}} = \frac{c_s \rho}{g} u_*^b \left[\frac{1}{u_*} - \frac{2u_{*t}}{u_*^2} - \frac{3u_{*t}^2}{u_*^3} \right] \tag{3.10}$$

CHAPTER 4

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APPENDIX A

ABBREVIATIONS AND ACRONYMS

APPENDIX B

SYMBOLS

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