Enhancements in tropospheric ozone and carbonaceous aerosols caused by Asian fires during Spring 2008 and the attendant radiative forcing

Murali Natarajan, R. Bradley Pierce, Todd Schaack, Allen Lenzen, Jassim Al-Saadi, Amber J. Soja, Thomas Charlock, and Fred Rose

Abstract

Biomass burning emissions cause perturbations in tropospheric ozone and carbonaceous aerosols both near and also far away from the fire locations. Significant changes in the abundance of these radiatively active components could have important implications for the regional scale radiative balance. Major outbreaks of fires occurred in three regions of Asia, namely Kazakhstan, Siberia, and Thailand, during Spring 2008. We have conducted simulations of the effects of these fires on atmospheric composition using the Real-time Air Quality Modeling System (RAQMS). RAQMS is a global scale meteorological and chemical modeling system with a unified chemistry module developed at NASA Langley Research Center, and a dynamical core developed at the University of Wisconsin. Results from these simulations, averaged over April 2008, indicate that tropospheric ozone column increases by more than 10 dobson units (DU) near the Thailand region mainly due to the effects of the fires. Widespread increases in the optical depths of organic and black carbon aerosols are also seen. Comparisons of the baseline model results with observations of tropospheric ozone and aerosol optical depth show reasonably good agreement. We have used an off-line radiative transfer model to evaluate the daily radiative flux profiles and also the radiative forcing due to the fire induced changes in composition. For clear sky conditions, the monthly averaged radiative forcing at the top of the atmosphere is mostly negative with peak values less than -12 W/m2 occurring near the fire regions. This negative forcing is mainly due to scattering of shortwave radiation by the increased aerosol loading. At high latitudes, the radiative forcing is positive with peak values of about 6 W/m2. This warming effect is caused by the presence of absorbing aerosols over regions of high surface albedo. Positive forcing at TOA occurs over wider regions for total sky conditions and this is again caused by the presence of absorbing black carbon aerosols over reflective cloud surfaces. The monthly averaged radiative forcing at the surface is mostly negative with peak values of less than -30 W/m2 near the fire regions. The contribution of increases in tropospheric ozone to the radiative forcing is positive and relatively smaller, but may not be insignificant on a regional scale depending on the location of the BB.

Introduction

Tropospheric ozone and carbonaceous aerosols are known forcing agents that affect the radiative balance of the Earth-atmosphere system (Forster et al. 2007). Their variability can impact the net radiative flux at the top of the atmosphere (TOA) and at the surface of the earth on a short time scale.

Biomass burning (BB) is one of the important drivers causing this variability. BB includes fires initiated for the purpose of clearing vegetation and changing land-use as well as uncontrolled wildfires. A major portion of BB is initiated by anthropogenic activities. Emissions from BB include Nitrogen oxides (NOx), Carbon monoxide, and non-methane hydrocarbons all of which are well known precursors leading to the formation of tropospheric ozone. BB is also a significant source of carbonaceous aerosols, contributing to about 76% of organic carbon (OC) and 42% of black carbon (BC) emitted globally (Bond, 2004).

Dominant greenhouse gases (GHG) including CO2 and CH4 have long lifetimes and they are well mixed in the atmosphere. As a result, they have long-term influence on global climate. In contrast, both ozone and carbonaceous aerosols have fairly short lifetimes in the troposphere. They are removed from the atmosphere over timescales of the order of days or weeks through wet and/or dry scavenging processes, and also, in the case of ozone, through chemical reactions. This short lifetime, combined with the inherent variability of the sources, results in spatial and temporal inhomogeneity in their distribution and radiative forcing. For example, local hot spots near fire zones experience large increases in aerosol optical depth, and photochemistry along the fire plumes results in enhanced production of tropospheric ozone. During such fire events, the radiative impact on a regional scale could be quite significant even though, when averaged over the globe, the effect maybe small. Increases in tropospheric ozone and absorbing aerosols like BC have a positive radiative forcing while the increases in scattering aerosols have a negative forcing. The potential for such short-term influences on regional scale radiative forcing has led to the suggestion that decreases in the emissions of absorbing aerosols could possibly alleviate the effect of global warming temporarily (Jacobson, 2002; Bond and Sun, 2005; Ramanathan and Carmichael, 2008). By the same argument, the presence of scattering aerosols may mitigate the effects of positive forcing due to long-lived GHG. Estimates of radiative forcing attributable to changes in the atmospheric abundance of different drivers are of interest, especially if control measures on anthropogenically initiated BB activities are to be considered.

Radiative effects of tropospheric ozone and carbonaceous aerosols have been explored in the past through combinations of global scale models and observations. Comparisons of the radiative forcing due to increases in tropospheric ozone relative to pre-industrial times are given in Gauss et al., (2006). These results are from various chemistry transport models (CTM) and coupled chemistry climate models. Globally and annually averaged change in tropospheric ozone column ranges from 7.9 to 13.8 DU. Corresponding direct radiative forcing (DRF) averaged over the models is 0.32 W/m2. The location and altitude of the ozone change have a strong influence on the radiative forcing. Joiner et al. (2009) make use of tropospheric ozone column derived from the Ozone Monitoring Instrument (OMI) and the Microwave Limb Sounder (MLS) experiments aboard the AURA satellite, and the radiative transfer code from the GEOS 5 DAS to calculate the radiative effect of tropospheric ozone. Their baseline case considers no tropospheric ozone and hence the calculated forcing represents an upper bound for the RF given in Forster et al. (2007). Naik et al. (2005) used the MOZART-2 model to evaluate the changes in tropospheric ozone due to a 10% reduction in the anthropogenic emission in 9 different regions, and calculated the radiative forcing using the GFDL radiative transfer model. They highlight the sensitivity of the ozone change and radiative forcing to season and geographical location of the perturbations in emissions. They also note that the radiative forcing is larger when the emissions of all the ozone precursors are reduced simultaneously. Martini et al., (2011) studied the impact of lightning and anthropogenic emissions on tropospheric ozone over North America. They find that the radiative forcing due to ozone generated from lightning NOx is slightly larger than that due to ozone produced from anthropogenic emissions.

In an earlier study, Hobbs et al (1997) used the optical parameters derived from airborne measurements in smoke from Brazilian fires, and estimated the global mean DRF due to aerosols from BB to be -0.3 W/m2. They also note that on a regional scale the radiative forcing due to smoke could be larger. Ross et al. (1998) evaluated the regional values of DRF from smoke aerosols generated by the Brazilian fires. They show that the DRF changes from -26 ± 6 W/m2 over a dark surface such as ocean to 25±12 W/m2 over a reflective surface such as a desert. In a case study of the solar radiative forcing by BB aerosols during SAFARI 2000 campaign, Keil and Haywood (2003) report the change of DRF at TOA from negative (cooling) to positive (warming) due to the presence of underlying cloud. Hsu et al (2003) used satellite data to investigate the impact of smoke aerosols on reflected shortwave and emitted longwave radiation from underlying cloud. They find that, in Southeast Asia during March 2000, the outgoing shortwave flux at TOA is reduced by as much as 100 W/m2 while the outgoing longwave flux is enhanced by about 20 W/m2 due to the presence of smoke aerosols over cloudy regions.

More recent studies have made use of global scale models to calculate the DRF of carbonaceous aerosols. For example, Naik et al. (2007), in a follow-up study, report the DRF due to regional reduction in BB emissions, which include OC and BC aerosols in addition to tropospheric ozone precursors. They conclude that a reduction in BB emissions results in negative RF at TOA and that the impact depends on the emission location. Chung et al. (2005) use a different approach involving MODIS, AERONET, and GOCART aerosol products and ISCCP cloud data to develop the aerosol distributions needed for radiative calculations. The anthropogenic aerosol RF is evaluated using the Monte-Carlo Aerosol Cloud Radiation model. They estimate a global mean forcing due to anthropogenic aerosols of -0.35 W/m2 at TOA, 3.0 W/m2 in the atmosphere, and -3.4 W/m2 at the surface. They note that regional differences in total surface forcing are large with tropical Asian regions displaying the largest contribution. They caution against heavy reliance on global mean forcing when assessing the role of aerosols. Wang et al., (2007) studied the regional climatic effects of BB emissions in Asia during the TRACE-P campaign time period. They consider a model domain spanning South and East Asia and use the HYbrid Single-Particle Lagrangian Integrated Transport (HYSPLIT) model to calculate the distributions of OC and BC aerosols during March 2001. The aerosol radiative forcing is calculated using the CLIRAD-SW model developed by Chou and Suarez(1999). Their results indicate a clear sky radiative forcing due to BB aerosols ranging from -1.9 to 0.4 W/m2 at TOA, and -0.5 to -12 W/m2 at the surface over the Asian region. The effects of tropospheric ozone changes are not included in the studies by Chung et al. (2005) and Wang et al. (2007).

In the present work, we focus on the radiative impact of the changes in tropospheric ozone and OC and BC aerosols triggered by the Asian fire events that occurred during the spring of 2008. Widespread fires occurred in 3 major areas of Asia, namely Thailand, Kazakhstan, and Siberia, over an extended period of the spring season. Figure 1 shows the three regions, labeled **A, B**, and **C,** in a composite of the locations of fires during April 10 to April 19, 2008, detected by MODIS (http://rapidfire.sci.gsfc.nasa.gov/firemaps/). The red color indicates low fire counts and the yellow color corresponds to high fire counts. The fires in the Kazakhstan region were mostly agricultural burning whereas those in the Lake Baikal region of Siberia were forest fires (Warneke et al., 2009). These uncontrolled wildfires exhibited large day-to-day variability. The transport of the smoke plumes from these fires across the Pacific and into the arctic region has been identified by satellite and aircraft observations and related analyses that were conducted during and after the ARCTAS and ARCPAC campaigns (Warneke et al., 2009, 2010; Dupont et al., 2010). We have used a 3-dimensional air quality model to simulate the atmospheric composition during the spring and summer of 2008 as a part of our support to the ARCTAS and ARCPAC campaigns. Additional sensitivity studies with and without the Asian fires are used in this study to evaluate the changes in tropospheric composition attributable to these fires. Radiative flux calculations, conducted using an offline radiative transfer model, help us evaluate the radiative impact of the fires. A description of the air quality model including the input data related to the fire events is given in the next section. This is followed by a discussion of the calculated species distributions from the baseline model and the changes caused by the Asian fire. In the next section, we present a brief description of the offline radiative transfer model and a discussion of the RF results. Summary and conclusions make up the last section.

Model Description

The main tool used for modeling the atmospheric chemical composition is the Real-time Air Quality Modeling System (RAQMS) developed jointly at the NASA Langley Research Center and the University of Wisconsin (LaRC/UW). The global scale meteorological and chemical modeling system has been used for assimilating remotely observed atmospheric chemical composition and forecasting the air quality within selected regions of the Earth (Pierce et al., 2003, 2007). The dynamical core for the global version of RAQMS is the UW hybrid isentropic coordinate model (Schaack et al., 2004). The tropospheric physics embedded in this model is the formulation used in the NCAR CCM3 model. Zapotocny et al. (1997) have demonstrated the veracity of the hybrid isentropic coordinate model in simulating the long-range transport of trace constituents. For the present study, we use the RAQMS model in a diagnostic (reanalysis) mode.

The simulations are conducted on a 2° x 2° horizontal resolution. The vertical domain extends from the surface to upper stratosphere. The meteorological fields are initialized with the NOAA Global Forecasting System (GFS) analysis every 6 hours. We include assimilation of the stratospheric ozone profile data from the MLS instrument aboard the AURA satellite. Assimilations of total ozone from the OMI experiment and aerosol optical depth from the MODIS experiment, which were done in support of ARCTAS campaign related studies, are turned off for the present study. This enables us to study the changes in tropospheric ozone and OC and BC aerosols caused by the Asian fires.

Photochemical mechanisms that control the formation and destruction of ozone in both the stratosphere and troposphere are included through a unified module. A family approach, which considers the continuity equations for the long-lived species and photochemical equilibrium relations for estimating the more reactive species, is adopted. The chemical kinetic scheme includes the standard Ox-HOx-NOx-ClOx-BrOx cycles that are important in the stratosphere, tropospheric NOx-HOx reactions, and the oxidation of CH4 and CO. Oxidation of non-methane hydrocarbons and isoprene is also included. A detailed description of the formulation is given in Pierce et al. (2007). The photochemical reaction rate data recommended by Sander et al. (2003) are used. Current version of FAST-J2, a code to evaluate the photolysis rate constants (Bian and Prather, 2002), is used. The photolysis rate constants are dependent on the calculated ozone vertical profile, aerosol optical depth and cloud parameters. RAQMS incorporates the Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model (Chin et al., 2002) to evaluate and apply the aerosol tendencies for settling, dry deposition, and chemistry. The input data for GOCART include calculated parameters such as OH concentration and horizontal wind. Aerosol types considered in the model are sulfate, BC, OC, dust particles of 4 different sizes, and sea salt (both coarse and accumulation modes). Both hydrophilic and hydrophobic types of carbonaceous aerosols are included. Tropospheric heterogeneous rates for the hydrolysis of N2O5 are based on model calculated sulfate distribution and properties.

Surface emissions of NOx and CO, other than those from BB, are based on climatological databases (GEIA/EDGAR). Data for Asian emissions have been updated based on Streets et al. (2003). NOx addition due to lightning and aircraft emissions are also included. Specification of BB emissions is of primary significance for the present study. These emissions invariably exhibit large spatial and temporal variability. To incorporate this variability adequately, RAQMS makes use of data from MODIS instrument aboard Terra and Aqua satellites in constructing a database of daily emissions from BB. The location, timing, and the area covered by the burning are obtained from the MODIS Rapid Response Fire Detection data. An eco-system dependent carbon fuel consumption database is used to estimate the carbon emissions under low, medium, and high fire weather severity conditions. Meteorology-based estimate of the fire weather severity is made using US Forrest Service Haines Index. The above data are combined to get the direct emission of carbon due to BB worldwide during spring and summer of 2008 (Al-Saadi et al., 2008). Separate estimates for day and night conditions are developed. Fluxes for different emitted species are evaluated using known emission ratios relative to carbon. Smoke injection height is parameterized based on the vegetation type and fire weather severity. The injections occur mostly within the planetary boundary layer and this is in accordance with the findings of Kahn et al. (2008).

Model Results and Evaluation

Past studies, through comparisons with available observations, have demonstrated the capability of RAQMS to simulate the impact of boreal fire plumes (Pierce et al., 2007, Verma et al. (2009), Dupont et al. 2010). These studies made use of TES observations of CO to identify the plumes. Backward and forward trajectories were used to confirm the origin of the plume, and through sampling of the RAQMS results for an ensemble of trajectories, the chemical evolution of the fire plume was examined. For the present study we employ the RAQMS model in a different manner. By conducting simulations with and without the BB emissions from the selected Asian regions, we generate baseline and perturbed model atmospheres so that the differences between them could be attributed to the Asian fires. We will first look at representative results from the baseline simulation (which includes the Asian fires) and comparisons with available observations.

Tropospheric aerosols

Ground–based optical monitoring aerosols has been conducted since 1993 as a part of the AErosol RObotic NETwork supported by NASA and various governmental agencies throughout the world (Holben et al., 1998). We have used the aerosol optical depth from AERONET locations obtained during April 2008 for a comparison with the results from RAQMS. A scatter-plot of the AERONET optical depth at 550 nm and the co-located RAQMS optical depth is shown in figure 2 (top left). The correlation coefficient is about 0.41 and the mean bias is 0.126 with RAQMS values lower than the AERONET data. In a study of atmospheric aerosol distribution from 2000 to 2007 using the GOCART model, Chin et al. (2009) report that the model underestimates the optical depth by 30-40% in the case of BB aerosols. The histogram of AOD at 550 nm given in the top right panel shows peak modes at lower values in AODs from RAQMS, which are not seen in the AERONET data. However, for AOD greater than 0.1, there is clear evidence of underestimation by the model, thus resulting in an overall negative bias. The bottom two panels of figure 2 show the correlation coefficient and the mean bias at the various measurement locations. We note that the AERONET coverage in the Asian fire regions is not as extensive as it is in Europe and North American continents. A few locations near the Thailand region show AOD model-data correlation coefficients greater than 0.5. The model AOD in some cases is lower than the data by as much as 0.5.

We have also utilized the global scale observations of aerosols obtained by the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument for comparisons with RAQMS model results. CALIOP, which is the main instrument aboard the Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations (CALIPSO) satellite, provides high-resolution vertical profiles and physical properties of clouds and aerosols (Winker et al., 2007). The satellite began its operations in 2006 as a part of the “A-Train” constellation. The observed total aerosol extinction, during the month of April 2008, is shown as a function of latitude and altitude in figure 3(a). Both day and night data from the longitude sector from 90E to 135E obtained under clear sky conditions have been used in generating this plot. The high aerosol extinction seen in the 10N to 50N latitude range and at low altitudes is most likely due to the influence of Asian wildfires that occurred throughout the month in this region. The vertical extent of this perturbation reaches nearly 5 km. Figure 3(b) shows the monthly averaged total aerosol extinction from the RAQMS baseline model for the same longitude sector. The model results have been interpolated to CALIOP measurement times and locations. The distribution looks quite similar to the CALIOP data but the RAQMS model seems to overestimate the extinction, especially near the surface and high latitudes. In a study of aerosol distributions using CALIPSO and MODIS observations from June 2006 to November 2007 and results from GOCART model simulations, Yu et al. (2010) report discrepancies in the magnitude of AOD in many regions. They show that the CALIOP extinctions are smaller than the GOCART values near the surface and in the free troposphere for most of the seasons over eastern China. While these conclusions are qualitatively similar to our findings, the reasons for these model-data differences are unclear.

Tropospheric Ozone

We make use of the ozone measurements made by the TES experiment for comparisons with the model derived ozone distributions in the troposphere during April 2008. The TES instrument was part of the payload aboard the AURA satellite launched in 2004. It is an infrared, Fourier transform spectrometer, which measures the thermal emission of the surface and the atmosphere. Vertical profiles of tropospheric ozone, CO, and various other trace gases are retrieved from this data on a global scale (Beer et al., 2001). Detailed description of the experiment and validation of the tropospheric ozone data are given in Worden et al. (2007). For the present study, we will focus on the data obtained during April 2008 in the longitudinal sector 90E to 135E and in the northern hemisphere. This longitudinal sector covers the fire zones of Thailand and Siberia. We first interpolate the RAQMS results to TES observation locations within the selected spatial domain and measurement time. Since the TES vertical resolution is coarser than the model vertical grid, we apply the TES averaging kernel (AK) to the ozone profiles from RAQMS. The resulting ozone distribution averaged over the month is shown in figure 4(a). The dark line at the top represents the average location of the model tropopause. Intrusions of stratospheric air mass are most likely responsible for mixing ratios higher than 150 ppbv indicated by yellow and red regions near the tropopause. Figure 4(b) shows the corresponding ozone distribution based on the TES observations. The agreement between the two distributions is quite good. Figure 4(c) displays the difference between the model (with TES averaging Kernel) and TES as a percentage of TES data. The model ozone (with TES AK) is generally within +/- 20% of the TES observations. Larger disagreement is seen near the equator suggesting that RAQMS is underestimating the convective transport of low marine boundary layer ozone into the upper troposphere. In the mid to upper troposphere near the Thailand fire zone (25N to 30N), the model ozone is lower, but is within 15% of the TES observations. Overall the agreement between the RAQMS and TES ozone data is quite good.

Figure 5 shows a comparison of the time series of tropospheric ozone from model runs with and without the influence of Asian fires, during April 2008. The location chosen for this comparison corresponds to the ozonesonde station at Naha, Japan (127.7 E, 26.2 N). The vertical dashed lines denote the 4 days when ozonesonde measurements were made at Naha. Large increases in ozone mixing ratio are noted in the left panel (with fire) compared to the right panel (without fire). The ozone perturbations occur even in the upper troposphere as seen, for example, around April 12 near 250 hPa. Comparisons of the ozonesonde observations and the calculated profiles corresponding to the 4 days of observations are shown in figure 6. The model values shown are at 6Z, which is within 30 minutes of the local time of measurements. The high values near 100 hPa most likely represent intrusions of stratospheric ozone. This is corroborated by figure 5, which shows not much difference at this level between the baseline and perturbed simulations. The observation on April 11 indicates a dramatic increase in the mixing ratio for the entire profile. Calculated O3 profile also shows an increase but the mixing ratios are lower than the observations. However, as seen in the time series (figure 5), the calculated O3 around 250 hPa shows even larger increases on April 12. We have studied the ozone sensitivity to individual fires by turning off emissions one region at a time. The results (not shown) indicate that the variability in calculated tropospheric ozone at Naha is mostly due to the fires in Thailand region.

The above comparisons demonstrate that the baseline RAQMS simulation, which includes emissions from all the BB, yields ozone and aerosol distributions which are in reasonable agreement with available data.

Impact of Asian Fires on Tropospheric Composition

The calculated changes in tropospheric composition that are attributable to the Asian fires are discussed in this section. We will focus on the changes in tropospheric ozone and carbonaceous aerosols only since they exert a major influence on the regional scale radiative balance. The results described here are the differences between the baseline simulation incorporating all the BB emissions and the perturbed case in which the emissions from the Asian fires (regions marked A, B, and C in figure 1) are excluded.

The change in tropospheric ozone integrated from the model surface to the tropopause for April 10 is shown in figure 7(top). The maximum value of about 24 DU occurs in the region A and values of the order of 10 DU are seen around this region extending into China. Large emissions of precursors like CO and NOx from the fire and the transport of the fire plume through regions known for high anthropogenic sources of pollution lead to enhanced production of ozone. The low latitude location is also conducive to more active photochemistry. In contrast, the high latitude fire regions of Kazakhstan and Siberia (regions B and C) do not show any significant increase in tropospheric ozone column. Increases in ozone column, though small, are seen around the globe at 30N. This is most likely due to the fires that occurred in region A prior to April 1. These plumes have had enough time to spread eastward across the Pacific. The bottom panels of figure 7 show the calculated changes in the BC and OC optical depths for April 10. The peak values of optical depths are collocated with the peak fire emissions in all the regions. The effect of transport is also noticeable in these maps. We note that this model considers all aerosols to be externally mixed. The optical properties and hence the optical depth could be different if internal mixing of two or more aerosols were allowed to occur.

Emissions from BB, especially those from wildfires, are characterized by large variability over short time scales. The ever-changing source emissions and atmospheric transport characteristics result in daily variations in the distributions of tropospheric ozone and carbonaceous aerosols. To illustrate this, we show the calculated changes for April 22 in figure 8. The top panel shows that the changes in tropospheric ozone column are more widespread but the peak values in region A are lower than on April 10. There is clear evidence of transport of ozone perturbations across the pacific with values of the order of 8 DU appearing near the international dateline and 20 N – 30N latitude range. The changes in BC and OC optical depths in the bottom panels show peak values that are larger than on April 10 in regions B and C. It should be noted that the MODIS fire count data for the region C peaked around April 17. Evidence for transport of the aerosols into the arctic region is also seen. Warneke et al. (2009) suggest that the earlier than usual onset of fire season in 2008 might have enhanced the efficiency of transport of BB emissions into the arctic region. The impact of the Asian fires on tropospheric composition averaged over the entire month of April is shown in figure 9. Ozone changes are predominant in the low latitude Thailand fire region with a peak value of the order of 10 DU. The BC and OC AOD changes are wide spread but the peak values are smaller than for a selected day shown earlier. The monthly averaged distributions provide a better view of the magnitude of the impact of the fires on a regional scale.

Radiative Transfer model

An objective of this study is to evaluate the radiative impact of the Asian fires, and we do this with the help of an off-line radiative transfer code. The model we use is a recent version of the NASA Langley Fu-Liou code. Shortwave calculation in this model considers 18 broadband and uses a 2-stream procedure. Longwave computation uses a combination of 2 and 4-stream approach (Charlock et al., 2006, Rose et al., 2006). H2O, CO2, O3, CH4, and N2O are the active species in the thermal infrared. The model uses correlated k distributions based on HITRAN 2000 in the shortwave computation. The model assumes an external mixture of gases, clouds and aerosols. Aerosol effects include scattering (SW and LW), absorption (SW and LW, and emission (LW). The aerosol properties are adopted from OPAC (Hess, 1998). Properties of water-soluble aerosols are used for OC and those of soot are used for BC. More information about this code and validations can be seen at the website of CERES/ARM Validation Experiment (CAVE) <http://www-cave.larc.nasa.gov/cave/>

Input data for the present calculations include daily averaged pressure, temperature, humidity, and ozone profiles from the RAQMS simulations. Also included are the daily averaged optical depth profiles for different types of aerosols from RAQMS. Cloud fraction, liquid water and ice concentration profiles, and direct and diffuse surface albedos are taken from the dynamical core (UW-Hybrid) model. Shortwave and longwave flux profiles at each grid point of the RAQMS model are evaluated for each day of April 2008. A Gaussian quadrature is used to get the diurnally averaged shortwave fluxes after repeating the flux calculations at the corresponding solar zenith angles. Net fluxes at the top of the atmosphere (TOA) and at the surface (SRF) are calculated for each day and grid point, for both the baseline and perturbed simulations. The difference between the net fluxes for the baseline (with fires) and the perturbed (without the fires) represents the radiative forcing due to the Asian fire emissions. It should be noted that we consider only the direct effect of the aerosols, namely those due to scattering and absorption. Cloud properties and surface albedo in the model are not affected by the changes in aerosol amount.

Radiative Forcing

The calculated impact of the Asian fires on the net radiative flux at TOA and SRF is discussed in this section. Just as we did earlier for the changes in composition, we will first look at the radiative effects for two separate days and then the monthly averaged results. Figure 10 shows the radiative impact for April 10. The top left panel corresponds to the results at TOA for clear sky conditions. Large negative forcing, lower than -20 W/m2 are seen near all the fire regions. This is mainly due to increased upward shortwave flux caused by aerosol scattering. The scattering effects of carbonaceous aerosols clearly dominate over the absorbing effects of BC for clear sky conditions. There is a hint of positive forcing in the arctic region at about 74 N and 135 E. The presence of absorbing BC aerosols above a reflecting surface (with high surface albedo) such as sea ice could result in such a warming effect. This is more clearly seen in the top right panel, which shows the radiative impact for total sky conditions. In this case, large negative forcing is still present close to the fire location. In region A, there is a sizeable area with positive forcing which is the result of enhancement of absorbing BC aerosols over the reflective cloud surface. The BC aerosols absorb the reflected shortwave radiation and warm the lower atmosphere. This reduction in the reflected shortwave at TOA shows up as a positive forcing. Hsu et al (2003) report a similar effect in their study of the modulating influence of smoke aerosols on the reflected radiation from clouds. Their conclusions were based on their analysis of narrowband radiances from SeaWiFS and TOMS and broadband data CERES taken in Southeast Asia during March 2000. Areas of positive forcing are also seen in the northern high latitudes of Asia, indicating the effects of the BC aerosols from the Kazakhstan and Siberian fires. We recognize that the magnitude of radiative forcing due to BC aerosols could be influenced by our assumption of an external mixing state for the aerosols. Jacobson (2001) reports that the mixing state of BC approaches that of an internal mixture and that this results in a higher positive forcing. The radiative forcing at the surface for the clear sky case is shown in the bottom left panel. Here, both the scattering and absorbing effects of the aerosols reduce the shortwave flux arriving at the surface and thus lead to a cooling of the surface. The minimum values are nearly -100 W/m2 in the Thailand fire region. The calculated impact at the surface is not much different for the total sky case shown in the bottom right panel. This is as expected since the cloud properties such as fractional cloud cover remain the same for both the baseline and perturbed model simulations.

Figure 11 shows the calculated radiative impact of the fires for April 22. At TOA for clear sky conditions large negative forcing is present very near the fire locations indicating the dominance of the aerosol scattering effects. Positive radiative forcing larger than 10 W/m2 is clearly seen in the arctic region. The influence of absorbing BC aerosols is further enhanced by the presence of clouds underneath as seen by the larger areas of positive forcing for the total sky case shown in the top right panel. Such warming influence over an extended period of time can certainly impact the fragile arctic climate. The bottom panels of figure 11 show the radiative forcing at the surface. For this day, the increase in aerosol-enhanced regions, shown in figure 8, translates into widespread cooling at the surface. Figure 12 shows the monthly averaged radiative forcing due to the emissions from the Asian fires. As in the case of changes in composition, the absolute peak values of the radiative forcing averaged over the month are much lower than for individual days. The peak forcing at TOA for clear sky case is about -20 W/m2 in the region A. Small positive forcing (3 W/m2 warming) is seen all over the arctic region. More regions of positive forcing are seen for the total sky case shown in the top right panel. The change from the clear sky case is quite significant in the regions affected by the Thailand fire. As expected, the monthly averaged forcing at the surface is negative in all the regions affected by the fire plumes.

In the above discussion of the radiative impact, we have highlighted mainly the role of scattering and absorbing aerosols. They certainly play a major role especially in the high latitude regions. The monthly averaged change in the composition, shown in figure 8, indicates that noticeable change in ozone occurs only in the low latitudes near region A. The radiative flux calculations that we have discussed do incorporate the effect of changes in ozone profiles. It is certainly of interest to look at the relative impact of changes in ozone alone. The radiative transfer model has an option to calculate the fluxes for a pristine atmosphere without any aerosol loading. Using this option, we have calculated the radiative forcing due only to changes in ozone. Since we are interested in the impact of changes in tropospheric ozone, it is better to look at net radiative fluxes at the tropopause level. Stratospheric ozone will modulate the TOA net flux and so it doesn’t show the full impact of tropospheric changes. Figure 13 shows the radiative forcing for clear sky conditions at the model tropopause. It should be noted that the color scale for the two panels are different because of the differences in the ranges of values. The left panel shows the monthly averaged combined effect of changes in aerosol optical depth and ozone. This is similar to the case shown in figure 12 except that the forcing is at the tropopause level. The forcing is mostly negative except in the high latitude and in the arctic region. The right panel shows the monthly averaged forcing due to changes in ozone alone. Positive forcing is seen in the low latitude affected by the fire in region A (Thailand). The maximum forcing near Laos is about 0.5 W/m2. We get similar results for total sky conditions (not shown). In the case of OC and BC aerosol enhancements, the flux changes are mostly in the shortwave. For O3 enhancements, shortwave effects are present but are minor compared to the longwave (greenhouse) effect. The reduction in upward longwave flux, both at TOA and at the tropopause, shows up as positive forcing.

We mentioned earlier that because of the short lifetimes of tropospheric ozone and carbonaceous aerosols, their impact on climate would be more significant on a regional scale than global scale. While the distributions shown earlier highlight the peak values and areal extent of the impact, they do not convey quantitative information for any region or for the globe. We have chosen the region A (Thailand and neighboring countries) to evaluate the regional impact of the fire. This region extends from 80E to 120E longitude, and from 10N to 30N latitude. The monthly averaged radiative impact for this region is shown in Table 1. The effects of changes in ozone and carbonaceous aerosol optical depth and those due to changes in ozone alone are shown in two major columns. The effect for clear sky is -2.546 W/m2 at TOA and -9.2 W/m2 at the surface when both ozone and aerosol changes are considered. This results in a net warming of the atmosphere. For total sky, the results are -0.54 W/m2 and -7.711 W/m2 at TOA and SFC respectively. Net warming of the atmosphere for total sky case is about 7.17 W/m2 compared to about 6.66 W/m2 for the clear sky. This difference is mainly due to the presence of absorbing aerosols above the reflective cloud cover. The radiative forcing due to changes in tropospheric ozone alone is 0.121 W/m2 at TOA and 0.145 W/m2 at the surface. The forcing at the tropopause level is 0.249 W/m2 for clear sky and this is larger than the TOA forcing. This difference is due to the modulating effect of stratospheric ozone. The forcing due to ozone alone at the tropopause for total sky conditions is similar in magnitude (but opposite in sign) to the forcing due to OC+BC and ozone. However, the warming of the troposphere by enhancements in BC aerosols caused by Thailand fires is more significant than that due to increase in ozone alone.

Summary

We have used a global scale air quality model and an of-line radiative transfer model to study the impact of emissions from Asian fires on the regional scale radiative forcing. This study considers the major fires that occurred in Kazakhstan, Siberia, and Thailand regions during Spring 2008. The main components of the emissions that are of significance to atmospheric radiative transfer are the carbonaceous aerosols (both OC and BC), and precursors for the formation of ozone. We made use of the MODIS rapid analysis data on fire counts along with meteorology based estimates of fire severity to develop day and night values of BB emission rates for the time period of interest. Our baseline case includes emissions from all the fires, and the perturbed simulation contains no emissions from the Asian fires. The difference between the results from the two model simulations indicates the extent of the perturbations in composition caused by the fire emissions. Increases in tropospheric column ozone of more than 10 DU occur in the low latitude region near the Thailand fire zone. Availability ozone precursors from other anthropogenic sources, combined with the faster photochemistry, in this low latitude region probably enhanced the formation of ozone. The ozone changes in the high latitude regions are not very significant, perhaps indicating the impact of slower photochemistry in the high latitudes during spring. Large increases in OC and BC optical depths are noticed near all the three fire zones. The day-to-day variations in BB emissions and in transport processes result in dramatic changes in the OC and BC distributions throughout the month. As shown in figure 8, by the 22nd of April, the aerosol perturbations have spread out to the arctic region. The impact of the boreal fire emissions on the arctic atmosphere was the focus of the ARCTAS and ARCPAC campaigns in 2008. But a large effect during the spring phase of the campaign was rather unexpected. Calculated aerosol distributions are qualitatively in agreement with the observations; however, discrepancies exist in the magnitude of AOD and extinction. Uncertainties in the emission source strengths used in the model could be partly responsible for these differences.

We use daily averages of the calculated distributions of ozone, OC, and BC, along with the other meteorological parameters from the RAQMS model as input data in an off-line radiative transfer model for evaluating the shortwave and longwave radiative flux distributions. The netflux values at the TOA, tropopause, and the surface from the baseline and perturbed model simulations are used to evaluate the radiative forcing due to the fire induced changes. Averaged over the entire month, the increase in the atmospheric reflectivity caused by the additional aerosol loading leads to a 20 W/m2 negative radiative forcing at the TOA near the fire zones under clear sky conditions. Away from the fire zones, and especially under cloudy conditions, positive radiative forcing is seen. This is mostly due to the transit of absorbing aerosol layer above the highly reflective cloud surface, which warms the atmosphere by absorbing the reflected SW. It is worth noting that, averaged over the entire month, a warming effect of about 3 W/m2 is seen in the arctic region far away from the fire regions, thus highlighting the importance of transport effects and the potential impact on the fragile arctic climate. At the surface, for both clear and total sky conditions, the predominant radiative forcing is negative. This is mainly due to the increased scattering and absorption of shortwave flux by the OC and BC aerosols. Given the short lifetimes of both tropospheric ozone and aerosols, the radiative effects of changes in their abundances are likely to be more significant on a regional basis. The monthly and area weighted average of the radiative forcing at TOA over the region A (80E to 120 E, 10N to 30N) and for clear sky conditions is -2.546 W/m2, and the forcing at the surface is -9.2 W/m2. This forcing includes the effect of changes in the tropospheric ozone within the region A. The contribution of changes in ozone alone, averaged over the month and over the region A, is 0.145 W/m2 at TOA and 0.245 W/m2 at the tropopause.

The radiative forcing results shown in this study are critically dependent on the input data and various parameterizations used in the model. This study focused mainly on the direct radiative effect of the OC and BC aerosols and tropospheric ozone generated by the Asian fires. The cloud parameters used in the baseline and perturbed cases are the same. Therefore, the influence of the clouds in modulating the aerosol effects is considered while the influence of the aerosols on the cloud properties is not. The calculated forcing also depends on the properties of the aerosols used in the model, and on the mixing state of the aerosols. Uncertainties in the evaluation of the fire emission data and the injection height of the smoke also affect the results of the study. Further advances in model formulations, quality of the input data, and the availability of observations for comparisons will improve our understanding of the regional climatic impact of BB emissions. These will also add credence to the calculated radiative forcing results, which maybe required for making policy decisions related to climate change.

References

Al-Saadi, J. A., et al., Intercomparison of near-real-time biomass burning emissions estimate constrained by satellite fire data, J. App. Rem. Sen., 2, 021504, 2008.

Beer, R., et al., Tropospheric emission spectrometer for the Earth Observing System’s Aura satellite, Appl. Optics, 40, 2356-2367, doi:10.1364/AO.40.002356, 2001.

Bian, H., and M. J. Prather, Fast-J2 accurate simulation of stratospheric photolysis in global chemical models, J. Atmos. Chem., 41, 281 – 296, 2002.

Bond, T. C., et al., A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi:10.1029/2003JD003697, 2004.

Bond, T. C., and H. Sun, Can reducing black carbon emissions counteract global warming?, Environ. Sci Technol., 39, 5921 – 5926, 2005.

Charlock, T. P., et al., The global surface and atmospheric radiation budget: An assessment of accuracy with 5 years of calculations and observations, Proceedings of 12th Conference on Atmospheric Radiation (AMS), Madison, Wisconsin, 2006.

Chin, M., et al., Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sun photometer measurements, J. Atmos. Sci., 59, 461 – 483, 2002.

Chin, M., et al., Light absorption by pollution, dust, and biomass burning aerosols: a global model study and evaluation with AERONET measurements, Ann. Geophys., 27, 3439-3464, 2009.

Chou, M. D., and M. J. Suarez, A shortwave radiation parameterization for atmospheric studies, in Technical Report Series on Global Modeling and Data Assimilation, 15, NASA/TM-1999-104606. 1999.

Chung, C., et al., Global anthropogenic aerosol direct forcing derived from satellite and ground-based observations, J. Geophys. Res., 110, D24207, doi:10.1029/2005JD006356, 2005.

Dupont, R., et al., Reconstructing ozone chemistry from Asian wild fires using models, satellite and aircraft measurements during the ARCTAS campaign, Atmos. Phys. Chem. Discuss., 10, 26751-26812, 2010.

Forster, P., et al., Changes in Atmospheric Constituents and in Radiative Forcing. In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

Gauss, M., et al., Radiative forcing since preindustrial times due to ozone change in the troposphere and lower stratosphere, Atmos. Chem. Phys., 6, 575 – 599, 2006.

Hess, M., P. Koepke, and I. Schult, Optical properties of aerosols and clouds: The software package OPAC, Bull. Am. Met. Soc., 79, 831 – 844, 1998.

Hobbs, P. V., et al., Direct radiative forcing by smoke from biomass burning, Science, 275, 1777-1778, 1997.

Holben, B. N., et al., AERONET – A federated instrument network and data archive for aerosol characterization, Rem. Sens. Environ., 66, 1-16, 1998.

Hsu, N. C., et al., Radiative impacts from biomass burning in the presence of clouds during boreal spring in southeast Asia, Geophys. Res. Lett., 30,5, 1224, doi:10.1029/2002GL016485, 2003.

Jacobson, M. Z., Strong radiative heating due to mixing state of black carbon in atmospheric aerosols, Nature, 409, 695-697, 2001.

Jacobson, M. Z., Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, J. Geophys. Res., 107, D19,4410, doi:10.1029/2001JD001376, 2002.

Joiner, J., et al., Accurate satellite-derived estimates of the tropospheric ozone impact on the global radiation budget,

Atmos. Chem. Phys., 9, 4447 – 4465, 2009.

Kahn, R. A., et al., Wildfire smoke injection heights: Two perspectives from space, Geophys. Res. Let., 35, L04809, doi:10.1029/2007GL032165, 2008.

Keil, A., and J.M. Haywood, Solar radiative forcing by biomass burning aerosol particles during SAFARI 2000: A case study based on measured aerosol and cloud properties, J. Geophys. Res., 108, (D13). 8467, doi:10.1029/2002JD002315, 2003.

Martini, M., et al., The impact of North American anthropogenic emissions and lightning on long-range transport of trace gases and their export from the continent during summers 2002 and 2004, J. Geophys. Res., 116, D07305, doi:10.1029/2010JD014305, 2011.

Naik, V., et al., Net radiative forcing due to changes in regional emissions of tropospheric ozone precursors, J. Geophys. Res., 110, D24306, doi:10.1029/2005JD005908, 2005.

Naik, V., et al., On the sensitivity of radiative forcing from biomass burning aerosols and ozone to emission location, Geophys. Res. Lett., 34, L03818, doi:10.1029/2006GL028149, 2007.

Pierce, R. B., et al., Regional Air Quality Modeling System (RAQMS) predictions of the tropospheric ozone budget over east Asia, J. Geophys. Res., 108, (D21), 8825, doi:10.1029/2002JD003176, 2003.

Pierce, R. B., et al., Chemical data assimilation estimates of continental U.S> ozone and nitrogen budgets during the Intercontinental Chemical Transport Experiment-North America, J. Geophys. Res., 112, D12S21, doi:10.1029/2006JD007722, 2007.

Ramanathan, V., and G. Carmichael, Global and regional climate changes due to black carbon, Nature Geoscience, 1, 221-227, 2008.

Rose, F., et al., CERES Proto-Edition 3 radiative transfer: Model tests and radiative closure over surface validation sites, Proceedings of 12th Conference on Atmospheric Radiation (AMS), Madison, Wisconsin, 2006.

Ross, J. L., and P. V. Hobbs, Radiative characteristics of regional hazes dominated by smoke from biomass burning in Brazil: Closure tests and direct radiative forcing, J. Geophys. Res., 103, D24, 31,925 – 31,941, 1998.

Sander, S. P., et al., Chemical kinetics and photochemical data for use in atmospheric studies, evaluation number 14, NASA Jet Propul. Lab., Calif. Inst. of Technol., Pasadena, CA, 2003.

Schaack, T. K., et al., Global climate simulation with the University of Wisconsin global hybrid isentropic coordinate model, J. Clim., 17, 2998- 3016, 2004.

Streets, D. G., et al., An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108(D21), 8809, doi:10.1029/2002JD003093, 2003.

Verma, S., et al., Ozone production in boreal fire smoke plumes using observations from the Tropospheric Emission Spectrometer and the Ozone Monitoring Instrument, J. Geophys. Res.,114, D02303, doi:10.1029/2008JD010108, 2009.

Wang, S. H., et al., Estimate of radiative forcing of Asian biomass-burning aerosols during the period of TRACE-P, J. Geophys. Res., 112, D10222, doi:10.1029/2006JD007564, 2007.

Warneke, C., et al., Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008, Geophys. Res. Lett., 36, L02813, doi:10.1029/2008GL036194, 2009.

Warneke, C., et al., An important contribution to springtime Arctic aerosol from biomass burning in Russia, Geophys. Res. Lett., 37, L01801, doi:10.1029/2009GL041816, 2010.

Winker, D. M., W. H. Hunt, and M. J. Mcgill, Initial performance assessment of CALIOP, Geophys. Res. Lett., 34, L19803, doi:10.1029/2007GL030135, 2007.

Worden, H. M., et al., Comparisons of Tropospheric Emission Spectrometer (TES) ozone profiles to ozonesondes: Methods and initial results, J. Geophys. Res., 112, D03309, doi:10.1029/2006JD007258, 2007.

Yu, H., et al., Global view of aerosol vertical distributions from CALIPSO lidar measurements and GOCART simulations: Regional and seasonal variations, J. Geophys. Res., 115, D00H30, doi: 10.1029/2009JD013364, 2010.

Zapotocny, T. H., A comparison of inert trace constituent transport between the University of Wisconsin isentropic-sigma model and the NCR community climate model, Mon. Weather Rev., 125,120-142, 1997.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | O3 + AOT | | O3 | |
|  | Clear | Total | Clear | Total |
| TOA | -2.546 | -0.540 | 0.121 | 0.120 |
| Tropopause | -2.473 | -0.457 | 0.249 | 0.218 |
| SFC | -9.204 | -7.711 | 0.145 | 0.077 |

Table 1. April 2008 mean radiative forcing (W/m2) due to fire emissions, averaged over the region from 80E to 120E, and from 10N to 30N