Ozone production in boreal fire smoke plumes using observations from the Tropospheric Emission Spectrometer and the Ozone Monitoring Instrument

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[1] We examine the photochemical processes governing the production of ozone in smoke from large Siberian fires that formed in July 2006 using colocated O3 and CO profiles as measured by the Tropospheric Emission Spectrometer as well as NO2 and aerosol optical depths as measured by the Ozone Monitoring Instrument. The Real-Time Air Quality Model (RAQMS) is used to explain the observed variations of O3. Enhanced levels of ozone up to 90 parts per billion (ppbv) are observed near and away from the Siberian fires (60°N and 100°E) when sunlight and NO2 are available. We also observe significantly low O3 amounts (less then 30 ppbv) in the smoke plume from Siberian fires in conjunction with optically thick aerosols. Despite this wide variance in observed ozone values, the mean ozone value for all observations of the smoke plume is close to background levels of approximately 55 ppbv in the free troposphere. Using RAQMS we show that optically thick aerosols in the smoke plume can substantially reduce the photochemical production of ozone and this can explain why the observed mean ozone amount for all plume observations is not much larger than background values of 55 ppbv. However, the anonymously low ozone amounts of 30 ppbv or less point toward other unresolved processes that reduce ozone below background levels in the plume.


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[2] Long-range transport of smoke emissions from boreal fires can increase the atmospheric abundance of pollution over population centers. Prior studies [Wotawa and Trainer, 2000; Forster et al., 2001] have shown, for example, that transport of emissions from the Canadian boreal fires can significantly increase atmospheric abundances of carbon monoxide (CO), aerosols, and ozone (O3) over North American and European population centers. Bertschi and Jaffé [2005] showed, using satellite observations of aerosols and global aerosol transport model, that Siberian fire emissions were the primary source of three air pollution events off the coast of Washington State in 2003.

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the plumes from these fires stretched across Siberia and the
Pacific Ocean. The availability of satellite observations of
CO, O₃, aerosols optical depth (AOD), and nitrogen dioxide
(NO₂), for this period provides a valuable opportunity to
evaluate our understanding of factors controlling boreal
fire emissions, their impact on atmospheric chemistry, and
the transport of the ozone produced in this plume globally.
This study examines O₃ produced and transported during
July 2006 Siberian fire smoke plumes as observed from
space-based observing platforms such as the Tropospheric
Emission Spectrometer (TES) and the Ozone Monitoring
Instrument (OMI) on the EOS-Aura satellite [Schoeberl et
al., 2006].

2. Satellite Observations Used in Analysis

2.1. TES

[5] The TES instrument is an infrared Fourier transform
spectrometer that measures the thermal emission of the
Earth’s surface and atmosphere over the spectral range
650–2250 cm⁻¹. It was designed to provide simultaneous
vertical profile retrievals of tropospheric O₃, CO and other
trace gases on a global basis [Beer et al., 2001; Beer, 2006].
The nadir footprint is 5.3 km across the spacecraft ground
track and 8.5 km along track for the 16-detector average
[Beer et al., 2001]. TES has two basic science operating
modes: Global Survey and Special Observations. Global
Surveys are conducted every other day while special obser-
vations are taken as needed in between Global Surveys. We
used global survey observations of TES O₃ and CO
obtained between 20 July and 12 August 2006 with a nadir
sampling of ~1.6° spacing along the ground track.

[6] The analysis presented here utilizes TES version 003
data [Osterman et al., 2005]. An overview of the TES
retention algorithm and error estimation are discussed by
Bowman et al. [2006] and the characterization of errors and
vertical information for individual TES profiles are dis-
cussed by Worden et al. [2004] and Kulawik et al. [2006].
The vertical resolution of TES nadir O₃ retrievals is about
6 km for cloud-free scenes, with sensitivity to both the
lower and upper troposphere [Worden et al., 2004; Bowman
e et al., 2006]. To date, TES tropospheric O₃ validation has
been conducted through comparisons with ozonesondes
[Worden et al., 2007] and lidar [Richards et al., 2008].
These validation studies show that TES O₃ estimates are
typically biased high in the upper troposphere by approx-
mately 10%. Nassar et al. [2007] shows that TES O₃ is
biased high by 3–10 ppb in the upper troposphere.

2.2. OMI

[7] OMI is an ultraviolet and visible nadir solar back-
scatter imaging spectrometer which provides nearly global
coverage in one day with a nadir spatial resolution of 13 ×
24 km². OMI measures solar irradiance and Earth radiances
in the wavelength range of 270 to 500 nm with a spectral
resolution of about 0.5 nm. These radiances are used for
estimating tropospheric column amounts of NO₂, HCHO,
SO₂, AOD, as well as total O₃ amount [Levett et al., 2006].

[8] The analysis in this study utilizes OMI aerosol optical
depth and NO₂ Level 2 version 3 data products. The OMI
Level 2 geolocated geophysical parameters (O₃, NO₂, SO₂,
BrO, HCHO, OCIO, Aerosol, and Cloud) data products are
at full instrument resolution, one orbit per file (http://
disc.sci.gsfc.nasa.gov/data/datapool/OMI/). The basic algo-
rithm for the retrieval of AOD and NO₂ from OMI data are
described by Torres et al. [2002], Boersma et al. [2007], and
Bucsel et al. [2006], respectively. The NO₂ product was
successfully validated with DC-8 aircraft NO₂ vertical
profiles [Boersma et al., 2008].

3. Modeling Tool Used in Analysis: Real-Time Air
Quality Modeling System

[9] Chemical and aerosol analyses from the Real-Time Air
Quality modeling System (RAQMS) and ensemble wild fire
trajectories are used to examine the different processes
influencing the evolution of trace gases (e.g., O₃ and CO)
within fire plumes during the 2006 Siberian boreal fires
event. RAQMS is a unified (stratosphere/troposphere),
online (meteorological, chemical, and aerosol) modeling
system which has been developed for assimilating satellite
observations of atmospheric chemical composition and pro-
viding real-time predictions of trace gas and aerosol distri-
butions [Pierce et al., 2003, 2007; Kittaka et al., 2004]. The
chemical formulation follows a family approach with parti-
tioning on the basis of photochemical equilibrium approx-
imations. The nonmethane hydrocarbon (NMHC) chemical
scheme is based on the carbon bond lumped structure
approach [Pierce et al., 2007]. Photolytic rates are calculated
using the FastI2 method [Bian et al., 2003]. The RAQMS
aerosol model incorporates online aerosol modules from
GOCART [Chin et al., 2002, 2003] and a sulfate-nitrate-
ammonium thermodynamic equilibrium model [Park et al.,
2004]. Nine aerosol species (SO4, NO3, NH4, hydrophobic
organic carbon (OC), hydrophilic OC, hydrophobic BC,
hydrophilic BC, dust, sea-salt) are transported. RAQMS
biomass burning emissions use twice daily ecosystem/severity
based emission estimates coupled with Moderate-Resolu-
tion Imaging Spectroradiometer (MODIS) Rapid Response
fire detections [Al-Saadi et al., 2008]. Total direct carbon
emissions are calculated as the product of area burned and
the ecosystem- and severity-specific carbon consumption
estimates. Ecosystem-dependent carbon consumption data-
bases for three classes of fire severity (low, medium, and
high) are considered. Fire weather severity is estimated
using the U.S. Forest Service Haines Index, which considers
atmospheric moisture and thermal stability [Haines, 1988].
Emissions of other species are determined by combining
published emission ratios for different ecosystems [Cofer et
al., 1991; Andree and Merlet, 2001]. During the chemical
and aerosol assimilation cycle the RAQMS meteorological
forecasts are reinitialized from NOAA Global Forecasting
System (GFS) analyses at 6 h intervals.

The RAQMS chemical analysis used in the current
study is from a retrospective 9-month (February–October
2006) 2 × 2° assimilation that includes assimilation of
cloud cleared OMI total column O3 measurements and O3
and CO profiles from TES nadir measurements. MODIS
onboard the Aqua satellite [Roper et al., 2005; Davies et al.,
2004] AOD was assimilated during the period from 15 to
31 July to provide observational constraints for the inves-
tigation of the influences of aerosol loading on Siberian
wild fire photochemistry. The MODIS AOD assimilation
cycle was initialized from a Global Modeling and Assimila-
tion Office (GMAO) aerosol forecast provided by Arlindo
da Silva (NASA/GSFC). For the wild fire case studies only
anthropogenic and biomass burning sources of carbona-
ceous aerosols were considered. Other aerosol species were
passively advected. During the MODIS assimilation cycle,
masses of all aerosol species were adjusted within each
model layer on the basis of the total AOD analysis incre-
ment and the relative contribution of each aerosol species to
the total layer extinction. The MODIS AOD compares well
with OMI AOD observations also used in this study with a
correlation of approximately 0.7 between the two data sets
but with OMI AOD showing slightly higher values than
MODIS of about 0.2 AOD [Ahn et al., 2008].

4. Methodology

During mid-July 2006, multiple forest fires were
recorded across the central Siberian Plateau between the
north-flowing Lena River and Lake Baikal along 95°E
longitude and 60°N latitude (http://earthobservatory.nasa.
gov/NaturalHazards/natural_hazards_v2.php3?img_id=
13728). The Spatial distribution of the Siberian fires
detected by MODIS during the period from 20 to 26 July
2006 is shown in Figure 1. The substantial fire hot spots
located over Siberia from the MODIS fire counts data
between 55 and 70°N are also shown in Figure 2 (marked
as red) on 24 July 2006, which was the day of the peak
Siberian wildfire emissions, as shown by MODIS fire
counts [Davies et al., 2004].

In order to examine the production of O3 within the
smoke plumes from these fires we first needed to identify
the plume signature. TES and OMI observations from mid-
July to mid-August 2006 were used. TES measurements
with CO abundances greater than 120 ppb [e.g., Wofsy et al.,
1992; McConnell et al., 1996] were identified as potentially
impacted by upwind boreal fires. Collocated AOD from
OMI were used as corroborating evidence. Images from
MODIS were acquired during this period to compare with
the satellite outputs as further corroborating evidence. We
used backward trajectories from these observations of
enhanced CO and aerosols to the fire source as well as
forward trajectories from the fire source using both the
FLEXPART model [Stohl et al., 1998] and RAQMS to
ensure that the observed air parcels came from the Siberian
fires of interest.

5. Discussions

5.1. Siberian Boreal Fire Emissions

The MODIS data revealed intense fire activity in the
Siberian region during July 2006, near 95°E and 65°N as
shown in Figures 1 and 2 (marked as red). Figure 2a shows
the FLEXPART 10-day forward trajectories from this fire
starting on 24 July 2006. The light blue lines represent the
forward trajectories for a plume starting from a TES orbit
around 98°E and 62°N with altitude levels between 0 and
15 km. Figures 2b, 2c, and 2d shows the FLEXPART 5-day
backward trajectories (light blue lines) starting from a TES
track location (shown as orange), corresponding to Figures 3,
4, and 6, respectively. The back trajectories in Figures 2b,
2c, and 2d suggests that air sampled on the selected TES
orbits is mainly from the Siberian fires as the trajectories
show significant recirculation in and around the fire source.
The forward trajectories in Figure 2a indicate large plumes
of smoke transported from northeastern Siberia to across the
Pacific during late July, providing opportunities for many
observations of the plume by TES and OMI.

5.2. TES and OMI Observations for Siberian Smoke

Plume

Because the smoke from the fires of interest travels
eastward and the Aura satellite travels along a polar orbit
there are many opportunities to observe cross section of the
fire plumes. We first show four examples showing vertical
cross sections of CO and O3 values across fire plumes. The
average CO and O3 mixing ratios is also presented along
these vertical cross sections. As discussed earlier, in order to
ensure that the observed air parcels are only related to
smoke from the fires, the CO values from each observation
are averaged together over the pressure (>400 hPa) range
for which CO values are larger than 120 parts per billion by volume (ppbv). The ozone values are also averaged using similar criteria.

[15] Figure 3a shows CO and O₃ profiles from TES on 24 July 2007 near the Siberian fire source (100–130°E). The location of the Siberian fires are observed by MODIS to be approximately between 90–110°E and 60–70°N (Figures 1 and 2). The vertical profiles of CO in Figure 3a shows values ranging from background levels (~80 ppb) to enhanced levels (between 120 and 250 ppb). Back trajectories from the regions of high CO (Figure 2b) indicate that observed air parcels have recirculated in and around the fire and therefore indicate the high CO is related to the fire emissions.

Figure 2. (a) The 10-day forward trajectories (blue lines) from peak Siberian wildfire emissions for a plume starting around 98°E and 62°N on 24 July 2006. (b–d) Five-day backward (blue lines) trajectories starting from a TES track (shown as orange), corresponds to Figures 3, 4, and 6, respectively. The trajectories represent the five different sets of latitude and longitudes pairs over the TES overpass and altitude levels between 0 and 15 km. Locations shown in red (marked as crosses) indicate the MODIS fire retrievals on 24 July 2006. The orbit tracks correspond to TES measurement locations chosen as examples in this study, as shown in Figures 4b, 6b, 3b, and 5b, respectively, starting from west to east.
Figure 3. (a) TES CO, O₃, and Averaging Kernel (AK) diagonals, (b) AOD and NO₂ tropospheric column amounts as observed from OMI, and (c) latitudinal variations of CO and O₃ mixing ratios averaged for CO >120 ppb and pressure >400 hPa for the plume values near the Siberian fires at 100–130°E on 24 July 2006. Overlaying the AOD and NO₂ (Figure 3b) is the TES orbit track (red curve) with the locations of the TES observations indicated by diamonds.
Figure 4. Same as in Figure 3 but for the plume away from the Siberian fires at 135–170°E on 31 July 2006.
AOD is moderately enhanced (AOD > 0.1) as well as back trajectories across the plume. The mean O3 and CO concentrations averaged over these same altitudes are shown in Figure 3c. The chemical evolution of the Siberian boreal fires is further explored by sampling the RAQMS chemical analysis on August 2006 where the air sampled originated from the Siberian smoke plumes. The mean O3 and CO is computed for each cross section along with the root mean square of the O3 variability across the plume. These values are shown for these cross sections in Figure 7. The symbols in Figure 7 show the mean values of O3 and O3 in the plume cross section and the bars show the RMS of the O3 in the cross section. We observe that O3 can vary significantly, with values ranging from 20 ppbv to 90 ppbv for a wide range of conditions. The sensitivity of the estimates depend on altitude. The sensitivity of the estimates at the indicated pressure level to the actual amount of the species at the same pressure level. The greatest sensitivity of the CO and O3 estimates is in the free troposphere between 400 hPa and 800 hPa. Peak values of the O3 and CO averaged over the averaging kernel correspond to peak values of O3 and CO, and the largest variations of O3 relative to background values of approximately 55 ppbv in the free troposphere. Consequently it is reasonable to assume that observed variations in O3 and CO amounts in the free troposphere are at similar altitudes.

In order to better examine O3 and CO variations within the plume we next average the CO values over those altitudes where CO is larger than 120 ppbv and the diagonal of the averaging kernel is larger than 0.05 and the sum of the diagonals (also known as the degrees of freedom for the averaging kernel matrices shown to the right of the CO and O3 estimates. The diagonal of the averaging kernel is the sensitivity of the O3 or CO estimate at the indicated pressure level to the actual amount of the species at the same pressure level. The greatest sensitivity of the CO and O3 estimates is in the free troposphere between 400 hPa and 800 hPa. Peak values of the O3 and CO averaging kernel diagonals correspond to peak values of O3 and CO, and the largest variations of O3 relative to background values of approximately 55 ppbv in the free troposphere. Consequently it is reasonable to assume that observed variations in O3 and CO amounts in the free troposphere are at similar altitudes.

[17] Figure 3b shows aerosol optical depths as observed by OMI between 100 and 130°E longitude and 50 and 75°N latitude. The TES orbit track is also shown as a red curve over AOD values and the location of the TES observations are shown as diamonds overlaying the orbit track. The aerosols provide corroborating evidence that the observed air parcel interacted with the boreal fire plume. In fact, the largest values of AOD between 100 and 110°E and 60–70°N correspond to the fire locations as seen by the MODIS fire count data in Figure 1. Back trajectories from the locations of the TES observations (Figure 2b) indicate that the air parcels observed by TES had intersected the location of the fire approximately three days earlier. Enhanced O3 is observed anywhere from 50°N to 74°N. NO2 values of approximately 2 × 1015 molec/cm2 and relatively low AOD (AOD < 0.5) as seen in Figure 3b, indicate the availability of O3 precursors and sunlight for photochemical production of O3.

[18] In order to better examine O3 and CO variations within the plume we next average the CO values over those altitudes where CO is larger than 120 ppbv and the diagonal of the averaging kernel is larger than 0.05 and the sum of the diagonals (also known as the degrees of freedom for signal) is larger than 0.5 for CO in the troposphere. The O3 concentrations averaged over these same altitudes are shown in Figure 3c for observations that meet the above criteria. The total error estimates, calculated using the total error for each profile, averaged over the selected altitudes [Worden et al., 2006], is also shown. We find the total error for these averaged quantities are about 8–12% for O3 and 6–10% for CO; these uncertainties are sufficient for resolving the observed O3 and CO variations. For this plume cross section, we observe values ranging from 130 to 180 ppbv for CO and approximately 35 to 90 ppbv for O3. There appears to be no correlation between the O3 and CO amounts.

[19] These same quantities are shown in Figure 4a for a region further away from the fire (approximately 50° in Longitude) and at a later date (31 July 2007). Enhanced levels of CO and AOD (AOD > 0.1) as well as back trajectories (Figure 2c) indicate the enhanced CO levels are due to the Siberian fire emissions. As seen in Figure 4c, enhanced O3 of up to 70 ppbv is observed in regions where CO is moderately enhanced (between 120 and 200 ppb) and AOD is moderately enhanced (AOD ~ 0.5) along with the availability of NO2. However, there are also a couple of observations with low ozone values which likely indicate that different parts of this large plume, covering up to 15° in latitude, have different chemical histories. We explore these low ozone values next.

[20] In contrast to the ozone shown in Figures 3 and 4, Figures 5a and 6a show cross sections of the fire plume where O3 is low relative to background values of approximately 55 ppbv in the free troposphere. Figure 5a and 5b shows CO, O3, AOD and NO2, respectively, directly over the region where the fires are burning on 24 July 2006. CO is observed with values exceeding 300 ppb (with the color scale on the top left panel saturating at 200 ppb) and the aerosol optical depth exceeding 4. NO2 is observed with values ranging between 1 × 1015 and 2 × 1015 molec/cm2. Ozone values ranging from 30 to 65 ppbv are observed. The lower ozone values occur in regions where AOD is optically thick (>3). Note that the high AOD values do not affect the TES estimates of CO and O3 because thermal infrared radiation is not absorbed much by aerosols produced in fire [Kirchstetter et al., 2004].

[21] This behavior of low O3 within a plume with significant aerosol optical depth and significantly enhanced CO amounts (greater the 200 ppb) is also observed (Figure 6a) four days later (on 28 July 2008) for a plume that is 40° away from the fire source at approximately 100°E. In both, the 24 and 28 July cases there is significant variation of O3 across the plume, with low O3 usually occurring in plume cross sections with high AOD (AOD > 3) (Figure 6b). Because this behavior is observed both over and away from the fire, where temperature conditions are expected to be different, we do not believe that incorrect estimates of temperature will affect these O3 retrieval results.

[22] To examine these relationships further we compute these same values as shown in Figures 4c through 8c for approximately 30 plume cross sections of mid-July–mid-August 2006 where the air sampled originated from the Siberian smoke plumes. The mean O3 and CO is computed for each cross section along with the root mean square of the O3 variability across the plume. These values are shown for these cross sections in Figure 7. The symbols in Figure 7 are the mean values of CO and O3 in the plume cross section and the bars show the RMS of the O3 in the cross section. We observe that O3 can vary significantly, with values ranging from 20 ppbv to 90 ppbv for a wide range of CO values. Despite this large variance, the mean O3 for all observations is about the same as the background value of about 55 ppbv observed at similar latitudes but without enhanced CO. This suggests that while enhanced O3 abundances are produced in this boreal fire plume, the average net O3 production in the plume is small which is consistent with prior observations of aged O3 plumes using tower and aircraft observations [Real et al., 2007; Mauzerall et al., 1996; Val Martin et al., 2006].

5.3. RAQMS Analysis

[23] The chemical evolution of the Siberian boreal fires is further explored by sampling the RAQMS chemical analysis along the ensemble fire trajectories to understand the different processes and time evolution of trace gases within the plumes, with a particular focus on the evolution of O3 and CO under different aerosol loadings. To investigate the
Figure 5. Same as in Figure 3 but for the plume near the Siberian fires at 80–110°E on 24 July 2006.
Figure 6. Same as in Figure 3 but for the plume away from the Siberian fires at 130–160°E on 28 July 2006.
impact of aerosols on O3 production rates within the wild
fire plumes we conducted two simulations, one with and one
without BC and OC aerosols in the photolysis calculations.

5.3.1. RAQMS Chemistry Run Without Aerosols

Figure 8 represents results from a photochemical
calculation of O3 in the RAQMS model in which an
ensemble of 10-day forward trajectories samples the model
output starting on 24 July 2006 for Siberia at 100°E, 60°N
location. This run was conducted for the chemistry only
simulation from RAQMS, that is, without aerosols in the
photolysis calculations. On the basis of the RAQMS wild
fire emission estimates, this time period is associated with
the highest wild fire emission rates for this Siberian fire.
The time evolution of O3 CO concentrations, NOy (or NOx
per day) and net O3 production rates (production-loss) is
shown in Figure 8 along the smoke plume from this 10-day
forward trajectory run. The time histories for each ensemble
member (dots) and ensemble mean (solid line) are shown.

There is a large increase in O3 concentrations in the fresh
part of plume (ensemble mean goes from 80 to 130 ppb
within the first 24 h). This increase in O3 concentrations
is due to large daytime net production of O3 (exceeding
120 ppb day 24 hrs after the start of the trajectory calcu-
lation) associated with high NO2 mixing ratios (net NO2

Figure 7. Scatterplot for CO and O3 mixing ratios for all
plume observations averaged for the Siberian fire period
(mid-July–mid-August 2006).

Figure 8. The 10-day forward trajectory simulation without aerosols initialized with 24 July 2006
upwind data and daily fire emissions (Tg/day) from real time MODIS fire counts at Siberia showing time
evolution of (a) O3, (b) CO concentrations, (c) NOy, and (d) O3 production rates.
emissions reach 34 ppb/day). The peak O₃ mixing ratios within the fresh wild fire plume in the model of nearly 140 ppb is significantly larger than observed by TES which shows peak values of 90 ppb in the vicinity of the wild fire plume (see Figure 3a).

5.3.2. RAQMS Sensitivity Run: Aerosols Impact on Ozone Production (High Aerosol Loading)

[25] A second ensemble of 10-day forward trajectories starting on 24 July 2006 Siberia is shown in Figure 9 for a RAQMS chemistry simulation that also includes BC and OC aerosols in the photolysis calculations. In contrast to the chemistry only simulation, the RAQMS model including aerosols shows only slight increases in O₃ concentrations in the fresh part of the plume with the ensemble mean ranging from 60 to 70 ppb within the first 24 hrs. This slight increase in O₃ concentrations is due to enhanced daytime net O₃ production (which was about 29 ppb/day at 24 hrs). This represents a fourfold decrease in net O₃ production relative to the model simulation without aerosols and suggests that optically thick aerosols (AOD > 3) during the most intense phase of the Siberian wild fire event significantly inhibit photolysis and hence greatly modifies the O₃ production; this conclusion is consistent with the results of Real et al. [2007] who also discussed the effects of aerosols on reduced O₃ production. However, neither simulation is able to replicate the low O₃ observations of about 30–40 ppb observed by TES, which may be due to other chemical processes at the fire source such as aerosol surface chemistry [e.g., Val Martin et al., 2006; Real et al., 2007] or O₃ destruction due to the titration of NOₓ within the fresh plume [Crutzen and Brühl, 2001].

6. Discussion and Conclusions

[26] Ozone production in the July 2006 Siberian boreal fire is examined using synchronous tropospheric observations of O₃ and CO from TES and observations of aerosol optical depth and NO₂ column abundances from OMI. These observations show that Siberian biomass burning emissions can produce elevated O₃ within the fire plume. However, O₃ abundances in the Siberian boreal forest fire plumes are highly variable, with some plumes showing O₃ enhancements of up to 90 ppb and others showing no enhancement or even O₃ depletion, with abundances of 30 ppb, much lower than background tropospheric values of about 55 ppb.

[27] We investigated the impact of aerosols on O₃ production rates within the wild fire plumes using the RAQMS...


Crutzen, P. J., and C. Brühl (2001), Catalysis by NOx as the main cause of the spring to fall stratospheric ozone decline in the Northern Hemisphere, J. Phys. Chem. A, 105(9), 1579–1582, doi:10.1021/jp001984h.


