Tropospheric Nitric Acid Columns from IASI Interpreted with a Chemical Transport Model

Matthew Cooper\textsuperscript{1}, Randall V. Martin\textsuperscript{1,2}, Catherine Wespès\textsuperscript{3}, Pierre-François Coheur\textsuperscript{3}, Cathy Clerbaux\textsuperscript{3,4}, Lee T. Murray\textsuperscript{5}

1. Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Nova Scotia, Canada.
2. Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, USA
3. Spectroscopie de l’Atmosphère, Chimie Quantique et Photophysique, Université Libre de Bruxelles, Brussels, Belgium
4. Sorbonne Universités, UPMC Univ. Paris 06; Université Versailles St-Quentin; CNRS/INSU, LATMOS-IPSL, Paris, France
5. NASA Goddard Institute for Space Studies and Lamont-Doherty Earth Observatory of Columbia University, New York, USA

Key Points
Tropospheric HNO\textsubscript{3} columns inferred from IASI with help of GEOS-Chem model
GEOS-Chem underestimates tropospheric HNO\textsubscript{3} in Southeast Asia
Treatment of lightning NO\textsubscript{x} is most likely contributor to model bias

Abstract
This paper interprets tropical tropospheric nitric acid columns from the IASI satellite instrument with a global chemical transport model (GEOS-Chem). GEOS-Chem columns generally agree with IASI over the tropical ocean to within 10\%. However the GEOS-Chem simulation underestimates IASI nitric acid over Southeast Asia by a factor of two. The regional HNO\textsubscript{3} bias is confirmed by comparing the GEOS-Chem simulation with additional satellite (HIRDLS, ACE-
FTS) and aircraft (PEM-Tropics A and PEM-West B) observations of the middle and upper troposphere. This bias is likely driven by the lightning NO$_x$ parameterization, both in terms of the magnitude of the NO$_x$ source and the ozone production efficiency of concentrated lightning NO$_x$ plumes. We tested a subgrid lightning plume parameterization and found that an ozone production efficiency of 15 mol/mol in lightning plumes over Southeast Asia would reduce the regional HNO$_3$ bias from 92% to 6%. Other sensitivity studies such as modified NO$_x$ yield per flash, increased altitude of lightning NO$_x$ emissions, or increased scavenging of HNO$_3$ required unrealistic changes to reduce the bias.

1. Introduction

Nitrogen oxides (NO$_x$ ≡ NO + NO$_2$) in the free troposphere largely control the production of ozone (O$_3$), an important greenhouse gas and atmospheric oxidant. The dominant sink for NO$_x$ is oxidation to nitric acid (HNO$_3$). HNO$_3$ is one of the main forms of reactive nitrogen (NO$_y$) in the free troposphere, representing up to 50% of NO$_y$ in the tropical upper troposphere [Kasibhatla et al., 1993; Folkins et al., 2006]. However, models generally have difficulty reproducing observed NO$_x$/HNO$_3$ ratios [Brunner et al., 2005; Singh et al., 2007]. Improved understanding of HNO$_3$ production and loss mechanisms can help to better constrain NO$_x$ emissions, and in turn improve understanding of ozone production and its effect on climate.

Direct measurements of free tropospheric HNO$_3$ are rare, particularly in the tropics. HNO$_3$ measurements taken during aircraft campaigns offer high precision but have limited spatial and temporal coverage. Satellite instruments are capable of providing superior temporal and spatial sampling in the tropics. HNO$_3$ concentrations have been retrieved from observations
from several satellite instruments, including LIMS [Gille et al., 1984], MIPAS [Tsidu et al., 2005], MLS [Santee et al., 2004], ACE-FTS [Wolff et al., 2008], and HIRDLS [Kinnison et al., 2008]. However these instruments are primarily focused on high latitudes or stratospheric altitudes. In this study we use the Infrared Atmospheric Sounding Interferometer (IASI) instrument on the MetOp satellite platform. IASI is a high resolution spectrometer that provides global observations of HNO₃ column abundances with an unprecedented spatial and temporal resolution [Wespès et al., 2009].

Lightning NOₓ has a large influence on tropospheric ozone as lightning NOₓ is emitted directly into the free troposphere where NOₓ lifetimes are longer [Liu et al., 1987; Pickering et al., 1990; Sauvage et al., 2007]. Best estimates of the global lightning NOₓ source range from 2-8 Tg N a⁻¹ but significant uncertainty remains in both the magnitude and vertical and horizontal distribution of the source [Martin et al., 2007; Schumann and Huntrieser, 2007]. A difficulty in modeling the effects of lightning NOₓ is that lightning is a sub-grid scale process which must be parameterized in chemical transport models. Lightning parameterizations are most often based on meteorological properties and are sensitive to the convection scheme used in the model [Tost et al., 2007; Koshak et al., 2013]. Lightning NOₓ production is often determined using a prescribed number of NOₓ molecules produced per flash, but this value varies significantly between models [Schumann and Huntrieser, 2007]. O₃ and HNO₃ concentrations are sensitive to the vertical placement of lightning NOₓ emissions [Labrador et al., 2005]. Uncertainty also arises when considering the dispersion rate of lightning NOₓ plumes as rates of chemical reactions vary nonlinearly with respect to NO concentrations [Lin et al., 1988]. Since HNO₃ is highly soluble and is quickly scavenged in convective updrafts, HNO₃ concentrations are sensitive to wet deposition in models [Giorgi and Chameides, 1986; Mari et al., 2000].
The following paper examines tropical tropospheric HNO$_3$ columns retrieved from IASI satellite measurements. Section 2 describes data sources and other tools used in this work. Section 3 describes GEOS-Chem, a state-of-the-science global chemical transport model that is used here to interpret the IASI observations. Section 4 describes how GEOS-Chem is used to investigate the ability of IASI to provide information about HNO$_3$ in the tropical troposphere. In the process of this evaluation a bias in the GEOS-Chem HNO$_3$ simulation over the tropical West Pacific and Indian Oceans is discovered. Section 5 discusses several possible methods for resolving the bias, including changes to wet deposition and lightning processes.

2. Observational Data

IASI was launched on the MetOp satellite in October 2006, into a polar sun-synchronous orbit with an equator crossing time of 9:30 AM and PM [Clerbaux et al., 2009]. IASI is a nadir viewing Fourier transform spectrometer measuring thermal infrared radiation between 645 to 2760 cm$^{-1}$. The IASI HNO$_3$ retrievals have ~1 degree of freedom of signal and thus provide a column HNO$_3$ observation with little vertical information [Hurtmans et al., 2012; Wespes et al., 2009]. IASI provides global coverage twice daily. Both day and night observations are used here. IASI scans across track either side of the nadir, with a total swath of around 2000 km. Each field of view is composed of four circular pixels of 12 km diameter at nadir. Evaluation of previous IASI HNO$_3$ columns indicates average errors ranging from 12% at middle to high latitudes up to 32% near the equator [Wespes et al., 2009]. Cloud information from the operational processing is used to reject observations with cloud coverage above 25%. HNO$_3$ columns for the year 2008 are used in this study.
HNO\textsubscript{3} observations from two other satellites are used for vertical profile information. The High Resolution Dynamics Limb Sounder (HIRDLS) on the Aqua satellite is a limb scanning IR filter radiometer launched in 2004 on a near polar sun-synchronous orbit \cite{Kinnison et al., 2008}. The vertical resolution in the tropical upper troposphere is approximately 1 km. Individual profile precision in the retrieval is 10-15\% but comparisons indicate a low bias of up to 30\% relative to ACE-FTS and MLS satellite HNO\textsubscript{3} observations \cite{Kinnison et al., 2008}. Observations from 2005 to 2008 are used here. The Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) is a solar occultation instrument measuring infrared radiation \cite{Bernath et al., 2005}. Its orbit is optimized for high latitudes and thus provides infrequent observations in the tropics, however complete coverage can be achieved if averaged over several years. Previous evaluation of ACE-FTS with aircraft measurements and the GEOS-Chem model indicates a high bias in HNO\textsubscript{3} of 15\% in the tropical upper troposphere \cite{Cooper et al., 2011}. Observations from 2004 to 2011 are used here.

We also use aircraft data from two of NASA’s Pacific Exploratory Mission campaigns to evaluate the HNO\textsubscript{3} simulation. The West Phase B (PEM-West B) consisted of 16 flights by the NASA DC-8 over the northwest Pacific Ocean in February-March 1994 \cite{Hoell et al., 1997}. The Tropics Phase A (PEM-Tropics A) also used the DC-8 aircraft for 17 flights throughout the tropical Pacific between New Zealand and Hawaii from August-September 1996 \cite{Hoell et al., 1999}. Individual HNO\textsubscript{3} measurement accuracy is reported as 30-35\% \cite{Hoell et al., 1999}.

Ozonesonde measurements at Kuala Lumpur (2.7° N, 101.7° E) are also used to evaluate the ozone simulation. These measurements are part of the Southern Hemisphere Additional Ozonesondes (SHADOZ, http://croc.gsfc.nasa.gov/shadoz/) network, a group of 16 ozonesonde
sites in the southern tropics [Thompson et al., 2003a, 2003b]. A total of 235 ozone profiles taken from the years 1998-2007 are used here.

3. GEOS-Chem

The GEOS-Chem global 3-D chemical transport model [Bey et al., 2001] version 9-01-03 (http://geos-chem.org) is used to interpret the IASI HNO$_3$ observations. GEOS-Chem is driven by assimilated meteorological data provided by the Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight Center. GEOS-5 meteorological fields for the year 2008 degraded to a horizontal resolution of 4°×5° are used here. The GEOS-Chem simulation has 47 vertical levels, extending from the surface to 0.1 hPa including approximately 35 levels in the troposphere. GEOS-5 employs the relaxed Arakawa-Schubert convective parameterization for shallow and deep moist convection [Moorthi and Suarez, 1992].

GEOS-Chem contains a detailed simulation of HO$_x$-NO$_x$-VOC-O$_3$-aerosol chemistry in the troposphere, including the most recent JPL/IUPAC recommendations as implemented into GEOS-Chem by Mao et al. [2010]. The global lightning NO$_x$ source of 6 Tg N a$^{-1}$ was constrained primarily using O$_3$ observations [Martin et al., 2002; 2007] which is consistent with the 5.5 ± 2.0 Tg N a$^{-1}$ used in current models [Stevenson et al., 2013] and more recent top-down estimates from satellite constraints of 6.3 ± 1.4 Tg N a$^{-1}$ [Miyazaki et al., 2014]. NO$_x$ is distributed within simulated deep convection in a manner consistent with satellite climatologies of lightning flashes as described by Murray et al. [2012]. Lightning NO$_x$ emissions are distributed relative to the cloud top height according to profiles based on aircraft observations and 3-D cloud-scale model simulations [Ott et al., 2010]. Anthropogenic NO$_x$ sources are from the Emission Database for Global Atmospheric Research (EDGAR) inventory [Olivier, 2001]
and overwritten by regional inventories in the northern midlatitudes. Biomass burning emissions are from the Global Fire Emissions Database (GFED v3) [van der Werf et al., 2010]. The stratospheric ozone simulation uses the Linoz algorithm of McLinden et al. [2000]. Monthly mean production rates and loss frequencies for other gases in the stratosphere are computed using archived data from the GMI model [Murray et al., 2013]. The wet deposition scheme used in GEOS-Chem is described in Liu et al. [2001] with updates to subgrid scavenging by Wang et al. [2013]. HNO₃ is highly soluble and is often completely scavenged within the convective updraft [Mari et al., 2000].

4. Interpreting IASI Tropospheric HNO₃ Columns and Results

A major challenge in inferring tropospheric HNO₃ columns from IASI observations is determining which portion of the column can be attributed to the troposphere. The coarse vertical sensitivity of IASI makes this difficult, as tropospheric HNO₃ features are smoothed in the retrieval such that they overlap into the stratosphere and vice versa.

In the comparisons that follow, GEOS-Chem profiles are smoothed using the IASI averaging kernels to simulate IASI’s coarse vertical resolution. This is done using the method developed by Rodgers and Conner [2003]:

\[ x'_{GC} \approx x_a + A(x_a - x_{GC}) \] (1)

where \( x_{GC} \) is the vertical HNO₃ profile from GEOS-Chem, \( x_a \) is the a priori profile used in the IASI retrievals, \( A \) is the IASI averaging kernel matrix, and \( x'_{GC} \) is the smoothed GEOS-Chem profile. The smoothed profile is then summed vertically to give a column value which can be
compared to columns retrieved from IASI. A similar method of smoothing GEOS-Chem with remote sensing instrument averaging kernels to examine tropospheric ozone was used by Zhang et al. [2010].

We use GEOS-Chem to aid the separation of the stratospheric and tropospheric columns. Over the remote tropical Pacific Ocean (20°S - 20°N, 140° - 180° W) tropospheric HNO₃ concentrations are small enough that the column can be assumed to be primarily stratospheric. We scaled the simulated stratospheric HNO₃ production and loss rates such that the total HNO₃ column over the remote Pacific Ocean matches the IASI column. Zonal symmetry in stratospheric HNO₃ concentrations then allows this scaling to be applied throughout the tropics. The smoothed stratospheric columns in GEOS-Chem are subsequently calculated by setting the simulated tropospheric concentrations to zero before smoothing the simulated profiles with the IASI averaging kernels:

$$x'_{GC, strat} \approx x_a + A(x_a - x_{GC(troposphere=0)})$$ (2)

The tropospheric columns are then calculated by subtracting the smoothed GEOS-Chem stratospheric column from both the smoothed model total column and the IASI column ($\Omega_{ASI}$):

$$\text{Smoothed GC Tropospheric Column} = \sum_{z=0}^{\infty} x'_{GC} - \sum_{z=0}^{\infty} x'_{GC, strat}$$ (3)

$$\text{IASI Tropospheric Column} = \Omega_{ASI} - \sum_{z=0}^{\infty} x'_{GC, strat}$$ (4)

where z is altitude.

Figure 1 shows tropospheric HNO₃ columns from IASI and GEOS-Chem. Emissivity features over deserts cause overestimations in the IASI columns [Wespès et al., 2009], and while this problem is largely confined to desert regions we have ignored all observations over land as a
precaution. GEOS-Chem is generally consistent with IASI over the tropics with a mean difference of 15% and a similar spatial distribution. Low concentrations over the Pacific Ocean and elevated concentrations over the tropical Atlantic are visible in both IASI and GEOS-Chem. However, IASI columns are up to twice as high as those in the simulation over the Indian Ocean and West Pacific Ocean regions (defined by 60°-160° E, 15°S-15°N, here referred to as Southeast Asia), and the Atlantic/Pacific contrast is stronger in the simulation. This bias exists throughout the year. As measurement errors in the IASI retrievals are not higher in this region than elsewhere in the tropics, the simulation is the likely source of the bias.

Figure 2 provides further evidence of a model bias through comparisons with additional satellite data. The left panel of Figure 2 shows the relative deviation from the tropical mean tropospheric HNO₃ columns near the Equator (10°S-10°N) as a function of longitude for IASI and GEOS-Chem. The middle panel shows this deviation calculated from the HIRDLS HNO₃ concentration at 163 hPa. The right panel uses ACE-FTS HNO₃ concentrations near 245 hPa. Since IASI gives column values and ACE-FTS and HIRDLS provide upper tropospheric concentrations, all three plots are normalized by their tropical mean values to facilitate comparisons, as well as to reduce the effect of instrument biases. All three plots show the familiar wave-one pattern of elevated concentrations over the tropical Atlantic (longitude range 0°-20° and 310°-360°). All three plots also show that GEOS-Chem underestimates HNO₃ over the Indian and West Pacific Oceans (longitudes 60°-160°E).

Figure 3 shows vertical HNO₃ profiles from the PEM-West B and PEM-Tropics A aircraft campaigns and from GEOS-Chem. The simulation is run using GMAO MERRA reanalysis meteorological fields for the campaign years (1994 for PEM-West B, 1996 for PEM-Tropics A) and is sampled along the aircraft flight path. The simulation is consistently lower than
the aircraft measurements by a factor of 2-3 throughout the middle and upper troposphere. This confirms the bias in the simulation, as this is a consistent feature over a large vertical and horizontal range observed independently by satellite and aircraft measurements.

5. Understanding the GEOS-Chem Bias

In the following sections we examine the most likely sources of the model underestimate in Southeast Asia: the lightning NO\textsubscript{x} source and its subsequent chemical processes, and overly vigorous wet deposition.

5.1 Lightning NO\textsubscript{x} Yield per Flash

We first examine the magnitude of the lightning NO\textsubscript{x} source over Southeast Asia. In the year 2008 the standard GEOS-Chem simulation has a lightning NO\textsubscript{x} source of 6.2 Tg N a\textsuperscript{-1}. Figure 4 shows the effect of adding additional lightning NO\textsubscript{x} to the free troposphere. A factor of 3.5 increase in lightning NO\textsubscript{x} yield/flash over Southeast Asia yields a 2.4 Tg N a\textsuperscript{-1} increase in the total global annual mean lightning NO\textsubscript{x} source to 8.6 Tg N a\textsuperscript{-1}. The GEOS-Chem HNO\textsubscript{3} column bias versus IASI over the Indian Ocean and Indonesia is reduced from 92\% to 7\%. Agreement is also improved over the West Pacific. Figure 5 evaluates the implications for the O\textsubscript{3} simulation at Kuala Lumpur. The additional NO\textsubscript{x} leads to increased ozone production which causes errors of 25\% in the simulated ozone fields. This indicates that a simple increase in the lightning NO\textsubscript{x} source is not the ideal solution to the simulated column HNO\textsubscript{3} underestimate.

5.2 Vertical Distribution of Lightning NO\textsubscript{x}

The effects of lightning NO\textsubscript{x} on tropospheric chemistry depend on its vertical distribution, as NO\textsubscript{x} lifetimes and ozone production efficiencies generally increase with altitude
The profiles used to distribute lightning NO\textsubscript{x} emissions vertically developed by Ott et al. [2010] used information based on midlatitude and subtropical storms and might not be representative of tropical storms. We performed sensitivity studies by increasing the median injection height of tropical lightning NO emissions from 8.7 to 12.6 km. We find that raising the NO injection height increases the HNO\textsubscript{3} tropospheric column abundances throughout the tropics by as much as 25\% but has little effect in the regions where lightning NO is emitted as shown in Figure 4c. NO\textsubscript{x} emitted at higher altitude has a longer lifetime and can be transported horizontally to form HNO\textsubscript{3} away from the source region. This HNO\textsubscript{3} also has a longer lifetime at these altitudes leading to the increased column abundances throughout the tropics. However near the region of emission the increase in HNO\textsubscript{3} produced at higher altitudes is largely balanced by decreases at lower altitudes and does not significantly change the column abundance. Thus adjustments to the NO\textsubscript{x} injection height do not improve the simulation bias in Southeast Asia. The resulting change in ozone concentrations throughout the tropics is generally less than a few ppb as shown in Figure 5.

5.3 Subgrid Plume Parameterization

The method by which lightning NO\textsubscript{x} is emitted in the model may contribute to the model bias. Lightning NO\textsubscript{x} emitted into the GEOS-Chem grid boxes produce dilute NO plumes with typical concentrations less than 1 ppbv. Aircraft observations found that lightning can create highly concentrated NO plumes inside cumulonimbus clouds, with concentrations generally between 1-7 ppbv and occasionally rising as high as 25 ppbv [Huntrieser et al., 2002, 2009; Ott et al., 2010]. These concentrated plumes can have spatial scales as small as 300 m [Huntrieser et al., 2002] which are not resolved in global models with typical scales of 50-500 km. Ozone production from lightning NO\textsubscript{x} is highly nonlinear with respect to NO\textsubscript{x} concentration [Lin et al.,
As a result, the dilute NO\textsubscript{x} plumes created in GEOS-Chem overestimate the ozone production efficiency (OPE), or the number of ozone molecules produced per NO\textsubscript{x} molecule consumed. This means that O\textsubscript{3} is produced too efficiently and HNO\textsubscript{3} is produced too inefficiently. Similar issues have been noted in the representation of plumes from power plants [Sillman et al., 1990], aircraft [Meijer et al., 1997], and ships [Vinken et al., 2011]. Changes to the lightning NO\textsubscript{x} parameterization which account for this nonlinearity may reduce the simulated HNO\textsubscript{3} bias over Southeast Asia.

We explored this process by modifying the GEOS-Chem lightning simulation to account for the nonlinear chemistry that occurs in the early concentrated stage of a lightning NO\textsubscript{x} plume before it dilutes to grid box scale. This is done by allowing some of the lightning NO to rapidly convert to HNO\textsubscript{3} after a typical amount of ozone production. The ratio of O\textsubscript{3} to HNO\textsubscript{3} produced is treated as a constant OPE of 15 moles of O\textsubscript{3} per mole of HNO\textsubscript{3}. This value was estimated by forcing NO\textsubscript{x} concentrations in a GEOS-Chem grid box over Indonesia to 5 ppbv (near the middle of the measured 1-7 ppbv range for lightning plumes), calculating the instantaneous OPE and finding an average of 15 moles O\textsubscript{3}/mole NO. This value is similar to those found by modeling studies for urban pollution (5-20 mol/mol) [Kleinman et al., 2002] and for aircraft NO\textsubscript{x} emissions (10-28 mol/mol) [Gilmore et al., 2013].

Figure 4d shows the change in HNO\textsubscript{3} from adding globally 6 Tg N yr\textsuperscript{-1} with an OPE of 15 mol/mol. The lightning NO source was held fixed at 6 Tg N a\textsuperscript{-1} while the amount of extra N (emitted as HNO\textsubscript{3}) and O\textsubscript{3} was allowed to vary given uncertainty in plume dilution rates. The bias over Southeast Asia is reduced from 92\% to 9\%, but at the expense of increasing the bias throughout the rest of the tropics to -46\%. 
It is possible that lightning plumes over Southeast Asia have different behavior than elsewhere as lightning NO$_x$ yields are known to vary by region [Schumann and Huntrieser, 2007]. With this in mind, 0.5 Tg N is added with an OPE of 15 mol/mol over Southeast Asia only in Figure 4e. The bias over Southeast Asia is reduced to 6% without negatively affecting the rest of the tropics. Figure 5 evaluates how the additional O$_3$ (30 Tg) from this parameterization affects the O$_3$ simulation. Free tropospheric ozone concentrations in Southeast Asia increased by up to 7 ppbv (up to a 17% increase) with small increases elsewhere. Ozone in the middle troposphere remains within 25% of ozonesondes at Kuala Lumpur. The reasoning for treating lightning NO$_x$ over Southeast Asia differently than the rest of the globe is not fully understood at this time. Possible factors may be that flash radiances and thus LNO$_x$ productivity per flash may be higher near oceans than inland [Baker et al., 1999], or that intracloud lightning flashes, which dissipate more energy and may produce more NO$_x$, occur more frequently in the region [Corray 1997; Mackerras et al., 1998].

5.4 Wet Deposition

Wet deposition is another possible contributor to the model bias over this highly convective region. We explored the potential effect that deposition has on modeled HNO$_3$ by altering the HNO$_3$ scavenging efficiency. Figure 4f shows that reducing the scavenging efficiency in GEOS-Chem by a factor of two has little effect on IASI tropospheric columns, reducing columns over Southeast Asia and other convectively active regions by less than 10%. The reduction in solubility tested here is not physically likely but indicates that the convective scavenging scheme or errors in assimilated convection may play a role in the HNO$_3$ bias, although the overall sensitivity of the bias to scavenging is small compared to changes in lightning NO$_x$. 
6. Conclusions

We analyzed IASI tropospheric HNO$_3$ columns over the tropical ocean. IASI and GEOS-Chem tropospheric HNO$_3$ columns are consistent within 10% throughout most of the tropics. However, observations over Southeast Asia show column values twice as high as simulated values. This simulated bias was confirmed by aircraft measurements (PEM-West B and PEM-Tropics A) and observations from the ACE-FTS and HIRDLS satellite instruments.

Investigation into the source of the model bias indicates sensitivity to the lightning NO$_x$ parameterization. We found that direct changes to the lightning NO$_x$ source was unlikely to explain the bias as a large (factor of 3.5) increase was needed, which in turn led to a 25% bias in simulated ozone concentrations relative to ozonesonde observations. Studies examining the sensitivity of tropospheric HNO$_3$ columns to the vertical distribution of lightning NO$_x$ emissions showed that increasing the NO$_x$ injection height had little effect on the simulation bias, although led to increased column abundances by 25% away from the region of emission. A simple parameterization accounting for nonlinearities in the conversion of NO$_x$ to HNO$_3$ in the beginning stage of the lightning NO$_x$ plume and the related ozone production chemistry was implemented with moderate success. A prescribed subgrid ozone production efficiency of 15 mol/mol in conjunction with an additional 0.5 Tg N added over Southeast Asia reduced the bias in that region from 92% to 6% with minimal impact on simulated ozone concentrations.

We also found some sensitivity of the model bias to the HNO$_3$ wet deposition parameterization. Improved agreement between IASI and GEOS-Chem in convectively active regions was achieved by reducing the efficiency of wet scavenging in convective updrafts by a factor of two, although the overall effect was small. The most likely solution to the model bias
will include changes to multiple processes, although changes to lightning NO$_x$ may have a greater effect.

Future work should consider a more sophisticated lightning plume model in which the OPE depends on local dilution rates, perhaps similar to the parameterization for ship emissions [Vinken et al., 2011]. Additional processes not tested here, including errors in HNO$_3$ production from NO$_2$, HNO$_3$ photolysis in clouds, PAN chemistry, or uptake of HNO$_3$ by ice crystals [von Kuhlmann and Lawrence, 2006] may play a role.

Acknowledgements

IASI data are available on request by contacting P.-F Coheur. HIRDLS data products are available online at NASA’s Goddard Earth Sciences Data and Information Services Center. ACE-FTS data are available on request by contacting the ACE Science Team at info@scisat.ca. Aircraft data from PEM West and PEM-Tropics campaigns are made available online at NASA’s Global Tropospheric Experiment webpage at http://www-gte.larc.nasa.gov/gte fld.htm. SHADOZ ozonesonde measurements are available online at http://croc.gsfc.nasa.gov/shadoz/. Information on accessing GEOS-Chem code can be found online at geos-chem.org.

This work was supported by NSERC. P.-F. Coheur and C. Wespies are, respectively, Senior Research Associate and Postdoctoral Researcher with F.R.S.-FNRS. The research in Belgium was also funded by the Belgian State Federal Office for Scientific, Technical and Cultural Affairs and the European Space Agency (ESA Prodex IASI.Flow), as well as by EUMETSAT (O3MSAF).
References

Relationships between lightning activity and various thundercloud parameters: satellite and
modelling studies, *Atm. Res.* 51, 221-236

Bernath, P. F. et al. (2005), Atmospheric Chemistry Experiment (ACE): mission overview,

Bey, I., D. J. Jacob, R. M. Yantosca, J. A. Logan, B. Field, A. M. Fiore, Q. Li, H. Liu, L. J.
Mickley, and M. Schultz (2001), Global modeling of tropospheric chemistry with
assimilated meteorology: Model description and evaluation, *J. Geophys. Res.*, 106, 23,073-
23,096.

Brunner, D., Staehelin, J., Rogers, H. L., Köhler, M. O., Pyle, J. A., Hauglustaine, D. A.,
Jourdain, L., Berntsen, T. K., Gauss, M., Isaksen, I. S. A., Meijer, E., van Velthoven, P.,
Pitari, G., Mancini, E., Grewe, V., and Sausen, R.: An evaluation of the performance of
chemistry transport models - Part 2: Detailed comparison with two selected campaigns,

Clerbaux, C. et al. (2009), Monitoring of atmospheric composition using the thermal infrared

McLinden, D. A. Degenstein, A. Volz-Thomas, and C. Wesp (2011), Evaluation of
ACE-FTS and OSIRIS Satellite retrievals of ozone and nitric acid in the tropical upper
troposphere: Application to ozone production efficiency, *J. Geophys. Res.*, 116, D12306,


Liu, H., D.J. Jacob, I. Bey, and R.M. Yantosca (2001), Constraints from $^{210}$Pb and $^{7}$Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields, *J. Geophys. Res.* 101 (D11) 12109-12128.


Miyazaki, K., H.J. Eskes, K. Sudo and C. Zhang (2014), Global lightning NO\textsubscript{x} production estimated by an assimilation of multiple satellite data sets, Atmos. Chem. Phys., 14, 3277-3305, doi:10.5194/acp-14-3277-2014


van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T. (2010), Global fire emissions
and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009),
*Atmos. Chem. Phys.*, 10, 11707-11735, doi:10.5194/acp-10-11707-2010


Figure 1: Annual mean tropospheric HNO₃ columns for 2008 from IASI (top) and GEOS-Chem (middle). The difference between IASI and GEOS-Chem is also shown (bottom). The location of PEMWest-B and PEM-Tropics A flight paths used here are shown in black in the bottom panel.
Figure 2: Relative deviation from tropical mean HNO$_3$ over 10S-10N. The left panel contains GEOS-Chem and IASI tropospheric columns. The middle panel contains GEOS-Chem and HIRDLS mixing ratios at 163 hPa. The right panel contains GEOS-Chem and ACE-FTS mixing ratios at 245 hPa. All three panels show an underestimation in GEOS-Chem over Southeast Asia (shaded region spanning longitudes 60°-160°).
Figure 3: Average HNO$_3$ profiles from aircraft campaigns (PEM-West B and PEM-Tropics A) and from GEOS-Chem sampled along the flight paths. Error bars indicate one standard error from the mean.
Figure 4: Fractional difference in annual mean tropospheric HNO$_3$ columns between IASI and the following GEOS-Chem simulations: (a) Standard simulation, (b) with flash yield increased by factor of 3.5 (c) with median NO injection height increased from 8.7 to 12.6 km (d) with 6 Tg additional HNO$_3$ globally, (e) with 0.5 Tg additional HNO$_3$ over Southeast Asia only, and (f) with HNO$_3$ wet deposition efficiency reduced by 50%.
Figure 5: Annual mean ozone profile at Kuala Lumpur (2.7° N, 101.7° E). Profiles from a standard GEOS-Chem simulation (red), a simulation with 0.5 Tg HNO₃ added in Southeast Asia (blue), a simulation with the lightning NO source increased to 8.4 Tg (green), a simulation with lightning NO injection height raised to 12.6 km (pink), and an annual mean profile from ozonesondes (black) are shown. Error bars represent the standard error in the ozonesonde measurements.