### EOS Aura Science Team Meeting 30 August – 1 September, 2016 – Rotterdam, The Netherlands Oral Abstracts

### WACCM-D: modelling mesospheric ion chemistry for particle precipitation studies

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Energetic particle precipitation (EPP) affects the neutral chemistry of the middle atmosphere in the polar regions. For example, recent studies have shown that ozone variability caused by EPP can be tens of percent over solar cycle time scales. Ion chemistry (IC) plays a vital part in connecting EPP to the resulting changes in neutral odd hydrogen (HOx), odd nitrogen (NOx), and odd oxygen (Ox) species. Also, NOy partitioning can be modulated by ion chemistry, leading to increase of, e.g., nitric acid (HNO3) during EPP events. A good representation of ion chemistry is required to capture the observed changes in models. However, to date the complexity of the ion chemistry below the mesopause (i.e. in the D-region ionosphere) has restricted modellers to simplified ion chemistry parameterisations which are unable to reproduce some important effects, e.g. the increase of mesospheric HNO3. Here we use WACCM-D, a variant of the Whole Atmosphere Community Climate Model which includes a selected set of Dregion ion chemistry designed to produce the observed EPP/IC effects. We demonstrate the improved EPP/IC modelling by comparing WACCM-D results for the January 2005 solar proton event (SPE) to those from the standard WACCM and observations from the Aura/MLS and SCISAT/ACE-FTS instruments. The results indicate that WACCM-D significantly improves the modelling of HNO3, HCl, ClO, OH, and NOx during the SPE. Northern Hemispheric HNO3 from WACCM-D shows an increase by two orders of magnitude at 40-70 km compared to WACCM, reaching 2.6 ppby, in agreement with the observations. For HCl and ClO, the improvement is most pronounced in the Southern Hemisphere at 40-50 km where WACCM-D predicts decrease of HCl and increase of ClO by 1.6% and 10%, respectively, similar to MLS data. Compared to WACCM, WACCM-D produces 25-50% less OH and 30-130% more NOx at 60-85 km which leads to better agreement with the observation.

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Proposal for a new parameterisation of the instrumental spectral response function in DOAS retrievals and application to satellite measurements

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The instrumental spectral response function (ISRF) is a key quantity in spectroscopy. Within DOAS retrievals, the ISRF is needed for an accurate wavelength calibration and for the convolution of trace gas cross-sections to instrumental resolution. DOAS analysis

software like QDOAS or DOASIS allow the fitting of a high resolution solar spectrum to a measured spectrum based on a parameterized ISRF with predefined shape (e.g. Gauss, Lorentz, Voigt). For OMI, a more advanced ISRF ("broadened Gaussian") was determined which allows for flat-top and asymmetric ISRF; however, this ISRF model is computationally expensive due to the high number of parameters. Here we propose a "Super Gaussian" as further model function for the ISRF, which is similar to a Gaussian, but with the exponent k as additional free parameter:  $F(x) = A \exp(-(|x|/w)^k)$ . The parameter w determines the width of F, while k basically determines the shape. Optionally, different values for k and w can be allowed for the left vs. right branch of F to construct asymmetric ISRFs. This model function was found to be a good compromise between good fit results (i.e., F represents the actual ISRF much better than a Gaussian) for a wide range of tested ISRF shapes on the one hand, and robustness of the fit and low computation time on the other hand due to the low number of free parameters. A further advantage of this description of the ISRF is that the two partial derivatives, representing changes of shape and width, respectively, allow to mimic potential spectral structures caused by temporal changes of the ISRF (e.g. due to changes of the detector temperature) by adding pseudo-absorbers in the DOAS analysis. We investigate how far this affects different trace gas retrievals from satellite measurements.

# Top-down constraints on wetland CH4 emission variability from TES, SCIAMACHY, GOSAT and surface CH4 measurements.

A. Anthony Bloom (JPL/Caltech; abloom@jpl.nasa.gov), John Worden, Kevin Bowman, Meemong Lee.

Wetland methane (CH4) emissions account for 20–40% of the global CH¬4 budget. Large uncertainties in magnitude and timing of high-latitude and tropical CH¬4 dominate the global wetland CH4 uncertainty; these largely stem from poorly characterized role of process controls on wetland CH4 production. Multi-dataset CH4 constraints can potentially provide insights on the timing, magnitude and processes controlling the spatial and temporal variability of global wetland CH4 emissions. We use retrievals of lowertropospheric CH4 concentrations – based on SCIAMACHY, GOSAT and TES CH4 measurements – to confront an ensemble of wetland CH4 emission scenarios based on terrestrial biosphere models and wetland extent parameterizations. Our analysis is focused on key wetland regions (North American, Scandinavian and Siberian and Amazon wetlands) – in conjunction with the lower troposphere CH4 retrievals to produce revised regional estimates of mean wetland CH4 emissions. Based on our results and the ensemble of wetland CH4 emission scenarios, we place quantitative constraints on the role of temperature, carbon availability and inundation on the processes controlling global wetland CH4 emissions.

### Improved retrievals of NO2 from space: community best practices and quality assurance from the QA4ECV project

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We present a new, community best-effort algorithm for the retrieval of NO2 columns and apply it to spectra measured by the GOME, SCIAMACHY, GOME-2(A), and OMI sensors. The algorithm is based on the results from a thorough comparison and iteration of spectral fitting and air mass factor calculation approaches between IUP Bremen, BIRA, Max Planck Institute for Chemistry, and KNMI. These imply that liquid water absorption and an intensity offset should be included in the NO2 fitting for OMI in the 405-465 nm window, and that 425-450 nm is the preferred fitting window for morning sensors. The stratospheric NO2 column is determined from assimilating NO2 slant columns over remote regions in the Tracer Model 5 (TM5-MP) chemistry transport model. The representation of stratospheric NOy in the model is improved by nudging ODIN HNO3:O3 ratios, leading to more realistic NO2 concentrations in the free-running mode, which is relevant at high latitudes near the terminator. The coupling to TM5-MP also allows the calculation of air mass factors from a priori NO2 vertical profiles simulated at an improved spatial resolution of 1x1, so that hotspot gradients are better resolved in the a priori profile shapes. Other AMF improvements include the use of improved cloud information, a correction for photon scattering in a spherical atmosphere, and the inclusion of air mass factor uncertainty estimates in the final data product. We will show a comparison of our new OA4ECV retrieval product against existing data sets, highlight the more extensive quality assurance parameters now embedded in the product upon wide user request, and discuss the compatibility with the operational NO2 retrieval for the new S5P mission.

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#### Emergent constraints in chemistry-climate interactions: a Bayesian Approach

Kevin Bowman (JPL; kevin.bowman@jpl.nasa.gov), Noel Cressie, Le-Kuai, Helen Worden, Jeff Jewell

A putative objective of climate model evaluation is the reduction in uncertainty of climate model projections. However, the relationship between historic and present-day model performance and future model response is not straight-forward. So-called "emergent constraints" are a relatively new methodology that exploits correlations between present day performance of a climate model ensemble and the future response of that ensemble under climate forcing. We describe a new Bayesian approach that explicitly computes the future state distribution subject to present day observations. We apply this new methodology to observations of ozone and its radiative effect from the NASA Tropospheric Emission Spectrometer (TES) to constrain ozone radiative forcing

calculated from the Atmospheric Chemistry Climate Model Intercomparison (ACCMIP) ensembles. Based upon these emergent constraints, we quantify the bias in ACCMIP ensemble estimates of future ozone radiative forcing. We further show how model deviation implies different processes controlling ozone radiative forcing.

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# **Pollution over Megacity Regions from the Tropospheric Emission Spectrometer** (TES)

Karen Cady-Pereira (AER; kcadyper@aer.com), Jessica Neu, Vivienne Payne, Jennifer Hegarty, Ming Luo, Kevin Bowman

The JPL TES team has selected a number of megacities as Special Observation targets. These observations, or transects, are sets of 20 closely spaced (12 km apart) TES observations carried out every sixteen days. We will present the TES ozone (O3), peroxyacetyl nitrate (PAN), ammonia (NH3), formic acid (HCOOH) and methanol (CH3OH) data collected over Mexico City and Lagos (Nigeria) from 2013 through 2015. If time permits we will also present data over other cities in or near biomass burning regions, such as Delhi. We will combine the TES data with MODIS AOD and AIRS CO, as well as back trajectories, to determine to what degree the seasonality and spatial gradients in the TES measurements are driven by local emissions, biomass burning and regional circulation patterns. Some of the transects demonstrate very nicely the synergy obtained from simultaneous measurements of multiple trace species. We will also discuss how the spatial variability along the transects is related to topography and land use.

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#### First satellite observations of lightning-generated NOx in volcanic eruption clouds

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The generation of NO2 by lightning flashes is known to be an important source of NOx in the free troposphere, particularly in the tropics, with implications for ozone production. Although UV-visible satellite observations of lightning-generated NOx (LNOx) in thunderstorms have been previously reported, here we present the first satellite observations of LNOx generated by lightning in explosive volcanic eruption clouds (vLNOx) from the Ozone Monitoring Instrument (OMI) aboard NASA's Aura satellite. To date we have identified vLNOx in operational OMI NO2 measurements (OMNO2) during the high-latitude eruptions of Okmok (Aleutian Is; July 2008), Kasatochi (Aleutian Is; August 2008), Redoubt (Alaska; March 2009) and Grimsvötn (Iceland; May 2011). We have also detected vLNOx associated with eruptions of Ol Doinyo Lengai volcano (Tanzania) in March 2008, which is significant as this volcano produces little SO2 and hence its eruption clouds are otherwise difficult to detect. Analysis of OMNO2 data for other eruptions is underway. We use World Wide Lightning Location Network

(WWLLN) observations to verify the occurrence and location of lightning flashes in the volcanic eruption clouds. All the vLNOx anomalies are associated with strong UV Aerosol Index (UVAI) signals due to volcanic ash. Preliminary analysis shows that the maximum vLNOx column detected by OMI decreases linearly with time since eruption, and suggests that the vLNOx signal is transient and can be detected up to ~5-6 hours after an eruption. Detection of vLNOx is hence only possible for eruptions occurring a few hours before the daytime OMI overpass. To calculate more accurate vLNOx columns, we apply an algorithm designed to retrieve LNOx from OMI, which takes the total OMI slant column NO2 and removes the stratospheric contribution and tropospheric NO2 background and applies an appropriate air mass factor to convert the slant column LNO2 to a vertical column of LNOx. Based on the number of lightning flashes detected by WWLLN in each eruption cloud, we also estimate the vLNOx production efficiency (moles vLNOx per flash). Preliminary estimates for the 2008 Kasatochi eruption suggest that this is significantly higher than the production efficiency in thunderstorms, but may be biased high due to the low detection efficiency of WWLLN (< 10-50% of flashes detected over most regions). However, OMI measurements of LNOx in thunderstorms also suggest that any NOx below the cloud optical centroid pressure (OCP; ~350-500 hPa) is not detected, and we speculate that the OCP may be lower (i.e., at higher altitude) in fresh volcanic clouds due to higher optical depths. The observation of vLNOx in volcanic clouds is significant since it implies active convection and plume electrification close to the satellite overpass time, with implications for aviation hazards due to volcanic ash. Although vLNOx is undoubtedly a very minor fraction of global LNOx production, explosive volcanic eruptions may inject NOx into the stratosphere where it has implications for ozone chemistry.

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### Aerosol retrievals from the OMI 477 nm O2-O2 band - Focus over large urban area and cloud-free scenes

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The ability to monitor air quality and climate from UltraViolet-Visible (UV-Vis) satellite spectral measurements requires accurate trace gases (e.g. NO2, SO2, HCHO, O3) and aerosol observations. Aerosols and trace gases share, over urban and industrialized areas, similar anthropogenic sources, and their concentrations, as shown by the satellite observations, often present significant correlations (Veefkind et al., 2011). The importance of measuring atmospheric aerosols and improving our knowledge on their properties on a global scale is double. The first reason is their direct impact on the radiation budget of the Earth-atmosphere system through the scattering and absorption of solar and terrestrial radiation. The second reason is the large impacts of their uncertainties when retrieving vertical column densities of trace gases from UV-Vis air quality spaceborne sensors. Indeed, in the absence of clouds, the most important factor affecting the slant column observations of trace gases, and therefore the computation of related Air Mass Factor (AMF), is aerosols since their scattering and absorbing properties modify the

average light path distribution followed by the detected photons. In our study, we have investigated how aerosols impact the 477 nm O2-O2 spectral band of OMI [Chimot et al., 2016a] and then, how these impacts can help to retrieve Aerosol Optical Thickness and Aerosol Layer Pressure (ALP) for the first time over land. On the one hand, the OMI continuum reflectance at 475 nm is primarily constrained by the total column AOT of fine particles present in the observed scene as well as their optical properties. Indeed, reflectance values increase with increasing AOT independently of the ALP due to the additional scattering effects observed in the scene compared to an aerosol-free scene. On the other hand, aerosols at high altitude apply a strong shielding effect on the O2-O2 SCD: i.e. in the presence of aerosols, less photons are scattered back to the satellite sensor, and thus less absorption by O2-O2 is observed. The O2-O2 shielding effect by aerosols results from a combination of AOT and ALP. Multilayer Perceptron Neural Networks (NN) algorithms were then developed to retrieve Aerosol Optical Thickness (AOT) and Aerosol Layer Pressure (ALP) from OMI using the 477 nm O2-O2 band. These algorithms were applied for the first time over large urban and industrialized areas, in East China and cloud-free scenes over 3 years (2005-2007) [Chimot et al., 2016b, AMT manuscript to submit]. ALPs are retrieved with an accuracy of 400 m on average. However, such accuracy is obtained over scenes dominated by large AOTSs (> 1). Quality of the aerosol retrievals strongly rely to the assumptions about the aerosol model and the chosen surface albedo input. Potential expectations are allowed for a future explicit aerosol correction in the retrieval of trace gases from UV-Vis satellite sensor. As an illustrative exercise, the new effects on tropospheric NO2 AMF, over scenes dominated by fine particles, will be illustrated and compared with current implicit effects considered through the OMI O2-O2 cloud algorithm [Chimot et al., 2016a; Veefkind et al., 2016].

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### Natural and anthropogenic aerosols in the UTLS: Sources and the role of Monsoon transport

Mian Chin (NASA/GSFC; mian.chin@nasa.gov), Tom Kucsera, Thomas Diehl, Huisheng Bian, Valentina Aquila, Qian Tan, Peter Colarco, John Burrows, Adam Bourassa, Landon Rieger, Douglas Degenstein, Jean- Paul Vernier, Bengt Martinsson

We present our study of decadal variations of UTLS aerosols in terms of the origins and transport mechanisms through modeling and analysis of observations. We use the global model GOCART, which incorporates emissions from anthropogenic, biomass burning, volcanic, and other natural sources including dust and sea salt, to simulate the aerosols and track their origins. The model results are compared to Aura and other satellite observations from OMI, MLS, CALIOP, OSIRIS, and Envisat instruments as well as aircraft observations from CARIBIC. Although volcanic sources exerts large, sporadical perturbation to the UTLS aerosol composition, mainly due to the nature of volcanic eruptions and relatively high altitude injections, anthropogenic aerosols, not only from Asia but also from other regions, are transported from surface to high altitudes mainly via the monsoonal convective transport with well-organized seasonal cycles in the UT

region. We estimate the relative contributions of natural and anthropogenic aerosols in different altitudes in UTLS and discuss the implication of the continuous increase of Asian anthropogenic emissions.

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### Improved space-based NOx emissions over remote regions

*Jieying Ding (Technology university of Delft / KNMI; ding@knmi.nl), Ronald van der A, Bas Mijling, Pieternel Levelt* 

We improve the emission estimate algorithm DECSO (Daily Emission estimates Constrained by Satellite Observations) to better detect NOx emissions over remote areas. We estimate total NOx emissions, which include biogenic emissions that often drive the seasonal cycle of the NOx emissions. We demonstrate the improvements implemented in DECSO for the domain of East Asia. The emissions derived by DECSO are in good agreement with other inventories like MEIC. In addition, the improved algorithm is able to better capture the seasonality of NOx emissions and it reveals ship tracks near the Chinese coasts that are otherwise hidden by the outflow of NO2 from the Chinese mainland. The precision of monthly emissions derived by DECSO v5 for each grid cell is about 20%.

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Aura's Ozone Garden: Despite 44% Decrease in OMI NO 2, Ozone –Induced Injury to Sensitive Plants is Still Very Evident

Bryan N. Duncan (NASA/GSFC; Bryan.N.Duncan@nasa.gov), John M. Skelly, Ginger J. Butcher

OMI NO 2 data indicate a 44% decrease in NO x in the Washington, DC area over the last decade and surface data indicate that ozone has decreased as well. However, bioindicator plants in Aura's Ozone Garden still show substantial ozone –induced symptoms year after year – the ozone air pollution problem has not been fully resolved. We will present data from the ozone garden at the Goddard Space Flight Visitor Center and discuss how the garden is an effective way to teach the general public about the continuing effects of tropospheric ozone pollution (and how it's different from ozone within the stratosphere), the current health risks of air pollution, and why it's important to monitor air pollution from space.

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### Ozone variability and tendencies in the upper troposphere and lower stratosphere based on Aura MLS and sonde data

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For over a decade, global daily ozone profiles from the upper troposphere to the mesosphere have been measured by the Microwave Limb Sounder on the Aura spacecraft. This dataset is explored with a focus on the tropical upper troposphere and lower stratosphere (UTLS). We compare the MLS ozone profiles to ozonesonde measurements, mainly from the Southern Hemisphere Additional Ozonesondes (SHADOZ) network and its continuing time series of reliable low latitude ozone profile records. We perform time series comparisons and multiple regression analyses, and we assess how well these two sets of ozone profiles agree, despite their differences in sampling and resolution. The multiple regression is based on a model with proxies that include quasi-biennial oscillation (QBO) and multivariate El Nino/Southern Oscillation (ENSO) index (MEI) terms, which represent significant known components of low frequency variability in ozone. We address the stability of the MLS ozone dataset and present estimates of the underlying linear trends in UTLS ozone for 2005-2015.

# MUlti-SpEctral, MUlti-SpEcies, MUlti-SatEllite (MUSES) retrieval algorithm for "A Train", Suomi-NPP, and TROPOMI Satellites

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Tropospheric ozone is at the juncture of air quality and climate. Ozone directly impacts human and plant health, and directly forces the climate system through absorption of thermal radiation. Carbon monoxide is a chemical precursor of greenhouse gases CO2 and tropospheric O3, and is also an ideal tracer of transport processes due to its medium life time (weeks to months). The Aqua-AIRS and Aura-OMI instruments in the NASA "A-Train", CrIS and OMPS instruments on the NOAA Suomi-NPP, IASI and GOME-2 on METOP and TROPOMI aboard the Sentinel 5 precursor (S5p) have the potential to provide the synoptic chemical and dynamical context for ozone necessary to quantify long-range transport at global scales and to provide an anchor to the near-term constellation of geostationary sounders: NASA TEMPO, ESA Sentinel 4, and the Korean GEMS. We introduce the JPL MUlti-SpEctral, MUlti-SpEcies, MUlti-SatEllite (MUSES) retrieval algorithm, which ingests panspectral observations across multiple platforms in a non-linear optimal estimation framework. MUSES incorporates advances in remote sensing science developed during the EOS-Aura era including rigorous error analysis diagnostics and observation operators needed for trend analysis, climate model evaluation, and data assimilation. Its performance has been demonstrated through

prototype studies for multi-satellite missions (AIRS, CrIS, TROPOMI, TES, OMI, and OMPS). We present joint tropospheric ozone retrievals from AIRS/OMI and CrIS/OMPS over global scales, and demonstrate the potential of joint carbon monoxide profiles from TROPOMI/CrIS. These results indicate that ozone can be retrieved with ~2 degrees of freedom for signal (dofs) in the troposphere, which is similar to TES. Joint CO profiles have dofs similar to the MOPITT multispectral retrieval but with higher spatial resolution and coverage. Consequently, multispectral retrievals show promise in providing continuity with NASA EOS observations and pave the way towards a new advanced atmospheric composition constellation.

# Latest advancements of Smithsonian Astrophysical Observatory OMI trace gas retrievals

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At SAO we produce formaldehyde, glyoxal, water vapor, bromine monoxide, and chlorine dioxide from OMI measurements. Our effort to improve the quality of these products continues with new developments. Here we present a summary of our work. Recent improvements of the formaldehyde retrieval have allowed us to completely remove long-term and unrealistic slant column trends. The long-standing issue of lacking air mass factor calculation error is now addressed by providing first order error estimate due to profile shape factors, surface albedo, cloud fraction, and cloud pressure. It is ready to be implemented in our next public release of formaldehyde, glyoxal, and water vapor retrievals. The most up-to-date version of de-striped glyoxal retrieval is being produced at NASA's SIPS servers. Details of the retrieval and de-striping strategy are provided along with early scientific results. Water vapor is the latest addition to our set of trace gas retrievals produced using OMI data. The water vapor data released at the Aura Validation Data Center have been validated. The water vapor retrieval algorithm has been updated with new improvements which result in better data quality for both land and ocean. For bromine monoxide, we have improved the treatment of surface characteristics in the calculation of air mass factors, and developed a simple algorithm to produce tropospheric vertical column densities outside polar regions.

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## Processes influencing global tropospheric ozone: insights from OMI ozone observations

# Lu Hu (Harvard University; luhu@seas.harvard.edu), Daniel Jacob. Xiong Liu, Patrick Kim, Lin Zhang

Here we use the GEOS-Chem chemical transport model as a platform to test our current knowledge of key factors controlling tropospheric ozone. We provide a comprehensive evaluation of the most recent version of GEOS-Chem model for global tropospheric ozone with an ensemble of observations, including 1) the worldwide ozonesonde observations from WOUDC and NOAA GMD, 2) MOZAIC-IAGOS observations of upper tropospheric ozone from commercial aircraft, and 3) OMI satellite observations of mid-tropospheric ozone. We demonstrate that OMI spatial and seasonal patterns are consistent with ozonesonde data for the recent years (2012-2013), suggesting the OMI instrument is stable over more than 10 years of operation since 2004. Such consistency between OMI and ozonesonde data allows us to apply OMI data to test the tropospheric ozone simulation at global scale particularly in the tropics and the southern hemisphere where in-situ observations are limited and sparse. Compared to OMI 700-400 hPa ozone observations, the current standard GEOS-Chem simulation shows no global bias but distinct regional and seasonal patterns. The improved ability to model ozone compared to early model versions reflects the advancing scientific knowledge of atmospheric processes controlling global tropospheric ozone. We discuss its implications with a particular focus on recent model developments in meteorological input, lightning, isoprene chemistry, and tropospheric bromine chemistry.

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### Validation of 10-year SAO OMI Ozone Profile (PROFOZ) Product Using Ozonesonde and Microwave Limb Souder (MLS) Observations

Guanyu Huang (Harvard-Smithsonian Center for Astrophysics; guanyu.huang@cfa.harvard.edu), Xiong Liu, Kelly Chance, Kai Yang, P.K. Bhartia, Zhaonan Cai

We validate the Ozone Monitoring Instrument (OMI) ozone profile (PROFOZ) product from October 2004 through December 2014 retrieved by the Smithsonian Astrophysical Observatory (SAO) algorithm against ozonesonde and Microwave Limb Sounder (MLS) observations. We also evaluate the effects of OMI Row anomaly (RA) on the retrieval by dividing the data set into before and after the occurrence of serious OMI RA, i.e., pre-RA (2004-2008) and post-RA (2009-2014). The retrieval shows good agreement with ozonesondes in the tropics and mid-latitudes and above ~20 km in the high latitudes, and it demonstrates clear improvement over the a priori down to the lower troposphere in the tropics, and down to ~5/10 km on average at middle/high latitudes. The mean biases (MBs) of stratospheric ozone column (SOC) are within ~3.0 DU (2%) with standard deviations (SDs) of < ~10 DU (5%), and the MBs of tropospheric ozone column (TOC) are within 1.5 DU (6%) with SDs of < ~5.0 DU (15%). The comparison generally degrades at larger solar zenith angle (SZA) due to weaker signal and more subject to sources of errors, leading to worse performance at high latitudes and during the midlatitude winter. It also degrades with increasing cloudiness below ~15 km, and varies with cross-track position, especially with large MBs and SDs at extreme off-nadir positions. In the tropics and mid-latitudes, the post-RA comparison is significantly worse with larger SDs of up to 2% in the stratosphere and 8% in the troposphere and up to 6% in TOC, and shows systematic differences varying with latitude compared to the pre-RA comparison. The retrieval comparison demonstrates good long-term stability during the pre-RA period, but exhibits statistically significant trend of 0.14-0.7%/year above ~17 km, 0.7 DU/year in SOC and -0.33 DU/year in TOC during the post-RA period. Similarly, we compare the OMI stratospheric ozone profiles and SOC with collocated MLS data. The global mean biases are within 5% between 0.4hPa and 100hPa, 5-10% below 100hPa and ~8% above 0.4hPa. The SDs are 6-25% between 0.4-50hPa, 18-25% below 100hPa and ~22% above 0.4hPa. OMI profile shows latitude- and SZA- dependent biases and the comparisons generally degrade with the increase of SZAs and latitudes. These MBs are mostly within 8% and the SDs are mostly within 15% except in low altitudes and high latitudes. The SOC215 (stratospheric ozone column above 215hPa) shows a small bias of 0.51% with a SD of 2.76%. When compared as a function of latitude and SZA, the MBs are within 4.5% and the SDs range from 1 to 4%. The profile comparisons indicate that the post-RA profile comparisons are worse with larger SDs and MBs in high altitudes and northern high latitudes than the pre-RA comparisons. Furthermore, the MBs and SDs of SOC215 comparisons decrease from 0.18% and 2.29% in the pre-RA to 0.90% and 3.19% in the post-RA, respectively. The comparisons show a good long-term stability in the pre-RA period. However, SOC215 comparisons indicate ~0.2 DU/year trends over the globe except for the S60°-S90° region. The spatiotemporal variation of the retrieval performance suggests the need to improve OMI's radiometric calibration especially during the post-RA period to maintain the long-term stability and reduce the latitude/season/SZA and cross-track dependence of the retrieval quality.

# Exploring relationships between land conditions and air quality using models, satellite and aircraft observations

Min Huang (George Mason University/now visiting U of Maryland; minhuang@umd.edun), many from Aura, ARCTAS and recent NASA NCA and SUSMAP projects

Weather-dependent natural emissions, such as soil dust and biogenic volatile organic compounds (VOCs), are critical contributors to background pollutants. We explore the relationships between land conditions and air quality during spring-summer 2008 and 2015-2016 over the Northern Hemisphere, using various modeling and observational datasets. A number of WRF simulations were conducted using different land surface models, land cover data, and soil moisture initialization on a 60x60 km horizontal resolution grid over the Northern Hemisphere (as defined in Huang et al., 2010). The

modeled land and atmospheric states in multiple cases will be evaluated against satellite, surface and aircraft observations, as well as independent modeling/assimilation products (e.g., NASA's Global Land Data Assimilation System products). We then compute dust and biogenic emissions based on weather fields (e.g., 2m temperature, solar radiation, soil moisture, precipitation, winds) from these simulations. Results will be presented separately for (semi-) arid and vegetated regions, in terms of: 1) the temporal variability of dust and VOC in relation to the weather conditions; and 2) the sensitivity of calculated dust and VOC emissions to the used WRF fields: Over erodible areas defined by satellite land products, we focus on the linkages between observed dust (based on OMI absorbing aerosol index and MODIS deep blue aerosol products, as well as available aircraft and surface observations), the WRF-modeled and satellite soil moisture (i.e., from ASCAT, ESA CCI and NASA SMAP) and winds, as well as the calculated dust emissions; Over vegetated areas, we demonstrate the impact of weather conditions on the calculated biogenic VOC emissions, in connection with the aircraft isoprene measurements and the OMI formaldehyde data. Extended discussions will be given on temporal variability of biogenic VOCs and OMI nitrogen dioxide and their net effects on ozone under different chemical regimes.

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### ILMA: Applications of satellite observations of tropospheric NO2 at hIgh Latitudes for Monitoring Air quality

#### Iolanda Ialongo (Finnish Meteorological Institute; iolanda.ialongo@fmi.fi), J. Tamminen

Climate change scenarios foresee increasing anthropogenic activities and their environmental effects at high latitude regions (especially in the Arctic). In particular, nitrogen oxides (NOx = NO + NO2), mainly generated by combustion processes, play an important role in tropospheric chemistry and have harmful effects on human health. Satellite observations are best suited for atmospheric composition monitoring over large regions where ground-based measurements are only scarcely available or difficult to maintain. However, satellite observations are not yet extensively evaluated and applied for air quality monitoring at high latitudes. In this context, ILMA-project aims at evaluating the existing satellite retrievals at high latitude and increasing the scientific exploitation, with specific focus on NO2 observations in Finnish territory. NO2 are currently provided for example by OMI (Ozone Monitoring Instrument), operational onboard NASA's Aura satellite since 2004. The first results of the project show that OMI NO2 total columns and the ground-based observations from Pandora spectrometer in Helsinki (Finland) differ on average by -6% and 1% for all skies and clear sky conditions, respectively. The clear sky overpasses mainly correspond to summer days and, thus, to smaller solar zenith angles. The NO2 surface concentrations available from Kumpula air quality station in Helsinki are also analysed in order to evaluate the capability of satellite data to monitor air quality. The NO2 seasonal and weekly cycles from OMI, Pandora and surface concentration data show a similar wintertime peak and a lower signal during the weekend, as compared to the other weekdays. Also, OMI tropospheric columns in Helsinki show similar year-to-year variability over the last 10 years, as visible in the

surface NO2 concentrations. Also, the emissions from a recent volcanic eruption in Iceland affected air quality in Northern Finland, when the SO2 plume was transported eastward from the volcano. Also in this case, satellite SO2 observations (as available from OMI) showed similar spatio-temporal patterns as the ground-based SO2 surface concentrations from air quality stations in Finnish Lapland. These results add confidence in using satellite-based atmospheric observations for air quality monitoring at high latitudes. Future missions such as TROPOMI on Sentinel 5 Precursor will provide improved information on the atmospheric composition, increasing the possible exploitation and applications of such data at high latitudes.

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### **Observational Metrics for CMIP6 Climate Model Assessments**

*Jonathan H. Jiang (JPL/Caltech; Jonathan.H.jiang@jpl.nasa.gov)* 

Observational metrics based on satellite observations have been developed and effectively applied in the previous CMIP5 and post-CMIP5 model evaluation and improvement projects. As new physics and parameterizations continue to be included in models for the upcoming CMIP6, it is important to continue objective comparisons between observations and model results. This talk will summarize observational metrics and methodologies for constraining climate models with Aura and other A-Train satellite observations and support CMIP6 model assessments. We target parameters and processes related to atmospheric clouds and water vapor, which are critically important for Earth's radiative budget, climate feedbacks, and water and energy cycles, and thus reduce uncertainties in climate models.

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### Recent activities of the US OMI core team

Joanna Joiner (NASA GSFC; joanna.joiner@nasa.gov), David Haffner, PK Bhartia, Sergey Marchenko, Alexander Vasilkov, Wenhan Qin, Nick Krotkov, Brad Fisher, Robert Spurr, Chang-woo Ahn, Eun-Su Yang, Sungyeon Choi, Yasuko Yoshida

The US OMI core team provides general support to the KNMI instrument team (calibration and trend monitoring) and the international OMI science team in general. We will highlight some of the recent activities of the US OMI core team. The team is responsible for the maintenance and development of the OMI-TOMS total ozone product. Several new products have been recently developed by the core team that support scientific studies and algorithm development. These include various ancillary data sets collocated with the OMI footprints and a new angular-dependent Lambertian-equivalent reflectivity (LER) product based on MODIS-derived bidirectional reflectance distribution functions.

## Understand the hydrological controls on the tropospheric ozone greenhouse gas effect using AURA TES observations

Le Kuai (UCLA-Joint Institute for Regional Earth System Science and Engineering (JIFRESSE); lkuai@g.ucla.edu), Kevin W. Bowman, Helen M. Worden, Robert L. Herman, Susan S. Kulawik

The role of the hydrological cycle in controlling the greenhouse gas (GHG) effect of tropospheric ozone (O3) is quantified using the Aura Tropospheric Emission Spectrometer (TES) observations. We attribute the spatiotemporal variation of the TES O3 longwave radiative effect (LWRE), which is defined as the net reduction of top-ofatmosphere flux due to total tropospheric O3 absorption, to variations in relative humidity, surface temperature, and tropospheric O3 column. The maximum GHG effect for ozone, represented by LWRE, is found to be around 0.6 to 0.7 Wm-2 on zonal average in the subtropics. This maximum is related by low water vapor concentrations and suppression of clouds, which are driven by the downward branch of the Hadley cell over this region. Within the subtropics, the largest values of LWRE are over the Middle East (>1 W/m2) due to both large thermal contrast and tropospheric ozone enhancements from atmospheric circulation and pollution. Conversely, a lower ozone GHG effect (about 0.4 Wm-2 or lower) is found in the deep tropics closely following the Inter-Tropical Convergence Zone, attributable to strong water vapor absorption and clouds over deep convective regions. These results show that changes in the hydrological cycle due to climate change could impact the magnitude and distribution of ozone radiative forcing.

#### New Aura OMI NO2 standard product (Version 3)

Nickolay A. Krotkov (NASA GSFC; Nickolay.A.Krotkov@nasa.gov), E. A. Celarier, W. H. Swartz, S. V. Marchenko, L. N. Lamsal, E. J. Bucsela

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The Ozone Monitoring Instrument (OMI) onboard NASA's Aura satellite has been providing global observations of nitrogen dioxide (NO2) since October 2004. We describe a new version (V3) of the NO2 Standard Product (OMNO2) based on our sequential DOAS fitting algorithm. The new fitting algorithm results in 10%-35% lower slant column retrievals as compared to the version 2.1 product. This results in smaller stratospheric column amounts, in better agreement with independent measurements. We further improve the new data product by using monthly NO2 vertical profiles from a higher-resolution (1 degree latitude by 1.25 degree longitude) GMI chemistry and transport model (CTM) simulation with time-dependent NO2 emissions. Pending upgrades include the improved treatment of surface reflectivity, clouds, and aerosols.

# Effects of daily meteorology on satellite a priori and implications for constraint of NOx chemistry from space

Josh L. Laughner (University of California, Berkeley; jlaughner@berkeley.edu), A.H. Zare, R.C. Cohen

Satellite retrievals of tropospheric NO2 columns require modeling of assumed NO2 vertical profiles to properly account for the different sensitivity of the satellite to NO2 at different altitudes. Currently, no operational retrieval simulates these a priori profile at both high spatial and temporal resolution. We demonstrate that implementing high temporal resolution a priori NO2 profiles significantly alters the retrieved vertical column densities in a retrieval already using high spatial resolution profiles. For a case study around Atlanta, GA, individual column measurements change by up to 40% (2 x 10^15 molec. cm<sup>-2</sup>). Time averaged column measurements demonstrate a systematic change of up to 12%. These changes primarily result from correctly accounting for the change in NO2 distribution and lifetime resulting from different wind speeds and directions. Finally, we show that accounting for the fine spatial and temporal variations of NO2 in the a priori leads to a 50-100% increase in absolute NOx emissions derived from fitting wind aligned line densities.

# Recent reduction in NOx emissions over China: synthesis of satellite observations and emission inventories

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### Fei Liu (Royal Netherlands Meteorological Institute (KNMI); liu@knmi.nl), Qiang Zhang, Ronald J. van der A, Bo Zheng, Dan Tong, Liu Yan, Yixuan Zheng, Kebin He

Tropospheric nitrogen dioxide (NO2) column densities detected from space are widely used to infer trends in terrestrial nitrogen oxide (NOx) emissions. We study changes in NO2 column densities using the Ozone Monitoring Instrument (OMI) over China from 2005 to 2015 and compare them with the bottom-up inventory to examine NOx emission trends and their driving forces. From OMI measurements we detect the peak of NO2 column densities at a national level in the year 2011, with average NO2 column densities deceasing by 32% from 2011 to 2015 and corresponding to a simultaneous decline of 21% in bottom-up emission estimates. A significant variation in the peak year of NO2 column densities over regions is observed. Because of the reasonable agreement between the peak year of NO2 columns and the start of deployment of denitration devices, we conclude that power plants are the primary contributor to the NO2 decline, which is further supported by the emission reduction of 56% from the power sector in the bottomup emission inventory associated with the SCR penetration increasing from 18% to 86% during 2011–2015. However, a few urbanized regions (e.g., Beijing and Shanghai) reached the NO2 peak before the deployment of denitration devices for power plants, as a result of their strict regulations for vehicle emissions ahead of the national schedule.

### New-generation Aura OMI volcanic SO2 product: Algorithm description, initial results, and data continuation with S-NPP OMPS

### Can Li (UMD/ESSIC and NASA/GSFC; can.li@nasa.gov), Nickolay A. Krotkov, Simon Carn, Yan Zhang, Robert J. D. Spurr, Joanna Joiner

In June 2016, our group publicly released a new generation OMI volcanic SO2 product, the first major update to the product in  $\sim 10$  years. This upgrade is possible because of several advantages offered by our new principal component analysis (PCA) retrieval technique. In this presentation, we provide an overview of the new PCA-based OMI operational volcanic SO2 algorithm (OMSO2VOLCANO), with a focus on the comparison with the previous operational linear fit (LF) algorithm. To reduce retrieval noise and artifacts as seen in the LF product, the OMSO2VOLCANO algorithm uses characteristic features extracted directly from OMI radiances in the spectral fitting, thereby helping to minimize interferences from various geophysical processes (e.g., O3 absorption) and measurement details (e.g., wavelength shift). To reduce the low bias for large SO2 total columns in the LF product, the OMSO2VOLCANO algorithm employs a table lookup approach to estimate SO2 Jacobians (i.e., the instrument sensitivity to a perturbation in the SO2 column amount) and iteratively adjusts the spectral fitting window to exclude shorter wavelengths where the SO2 absorption signals are saturated. To the first order, the effects of clouds and aerosols are accounted for using a simple Lambertian equivalent reflectivity approach. As with the LF algorithm, OMSO2VOLCANO provides total column retrievals based on a set of pre-defined SO2 profiles from the lower troposphere to the lower stratosphere, including a new profile peaked at 13 km for plumes in the upper tropical troposphere. We will present a number of examples to compare the two algorithms under different scenarios, including both background cases and large eruptions. Overall, the OMSO2VOLCANO algorithm shows significant improvement over the LF algorithm, with at least 50% reduction in retrieval noise over the remote Pacific. For large eruptions such as Kasatochi in 2008 (~1700 kt total SO2) and Sierra Negra in 2005 (>1100 DU peak SO2), OMSO2VOLCANO generally agrees well with other algorithms that also utilize the full spectral content of satellite measurements, while the LF algorithm tends to underestimate SO2. We also demonstrate that, despite the coarser spatial and spectral resolution of the Suomi National Polar-orbiting Partnership (Suomi-NPP) Ozone Mapping and Profiler Suite (OMPS) instrument, application of the new PCA algorithm to OMPS data produces highly consistent retrievals between OMI and OMPS. The new PCA algorithm is therefore capable of continuing the volcanic SO2 data record well into the future using current and future hyperspectral UV satellite instruments.

### Structural uncertainty in air mass factor calculation process for NO2 and HCHO satellite retrievals

Alba Lorente (Wageningen University; alba.lorentedelgado@wur.nl), K. Folkert Boersma, Huan Yu, Steffen Dörner, Andreas Hilboll, Lok Lamsal, Mengyao Liu, Michael Barkley, Andreas Richter, Isabelle De Smedt, Michel Van Roozendael, Thomas Wagner, Jin Tai Lin

Air mass factor (AMF) calculation is the largest source of uncertainty in NO2 and HCHO satellite retrievals in scenarios with enhanced trace gas column concentrations. Structural uncertainty is one of the main sources of systematic uncertainty in satellite retrievals when multiple retrieval approaches might be applied to the same data. Here, we estimate the AMF structural uncertainty in every step of the AMF calculation process from a detailed evaluation and comparison of different AMF calculation approaches used by several scientific groups (KNMI/WUR, IASB-BIRA, IUP-Uni. Bremen, MPI-C, NASA GSFC, Leicester Uni. and Peking Uni) in their retrieval algorithms applied to measurements of the Ozone Monitoring Instrument (OMI). We evaluated top of atmosphere reflectances and altitude-dependent air mass factors computed with four radiative transfer models (RTMs) (DAK, McArtim, SCIATRAN and VLIDORT). Tropospheric AMFs were first computed using harmonized settings and afterwards using each group's preferred settings for the ancillary data (surface albedo, terrain height, and a priori profiles) and cloud and aerosol correction as part of a round robin exercise. We find that the choice of RTM introduces an uncertainty of 2-3% to the AMF calculation. The way Earth's sphericity is treated for multiple scattering in the RTMs contributes substantially to the altitude dependent AMFs differences for extreme satellite geometries in the troposphere and in the stratosphere. From the harmonized settings comparison, we estimated an AMF structural uncertainty of 6% due to interpolation errors and vertical discretization. From the round robin comparison, we estimated an overall structural uncertainty in the AMF calculation of 38% over polluted regions and 25% over unpolluted regions. Different cloud correction approaches result in substantial (5-40%) AMF differences even for low cloud fractions (0.05-0.2), and their magnitude depends mainly on cloud pressure. With the detailed round robin exercise, we showed that the choice of aerosol correction introduces an average uncertainty of 50% and we identified a priori trace gas profiles, surface albedo and cloud pressure as the forward model parameters that have the biggest effect on AMF structural uncertainty. We conclude that the assumptions and choices made to represent the state of the atmosphere add the largest structural uncertainty to the AMF calculation. Our work points to the need of a detailed validation exercise to evaluate different parameters in scenarios where AMF structural uncertainty has highest impact. The outcome of this study will be used in a community effort retrieval algorithm to be applied in past and future UV/Vis instrumentation in the European QA4ECV project.

# Solar Spectral Irradiance Variability in Cycle 24: Model Predictions and OMI Observations

Sergey V. Marchenko (SSAI; sergey\_marchenko@ssaihq.com), M. T. DeLand, J. L. Lean

Utilizing the excellent stability of the Ozone Monitoring Instrument (OMI), we characterize both short-term (solar rotation) and long-term (solar cycle) changes of the solar spectral irradiance (SSI) between 265-500 nm during the on-going Cycle 24. We supplement the OMI data with concurrent observations from the GOME-2 and SORCE instruments and find fair-to-excellent agreement between the observations and predictions of the NRLSSI2 and SATIRE-S models.

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#### Temperature-dependence of aerosol optical depth over the southeastern US

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Several studies have indicated that summer-time aerosol optical depths (AOD) over the southeastern US (70-90° W and 30-37.5° N) are dependent on temperature but the reason for this dependence and its radiative effects have so far been unclear. To quantify these effects we utilized AOD and land surface temperature (LST) products from the Advanced Along-Track Scanning Radiometer (AATSR) with observations of tropospheric nitrogen dioxide (NO2) column densities from the Ozone Monitoring Instrument (OMI). Furthermore, simulations of the global aerosol-climate model ECHAM-HAMMOZ were used to identify the possible processes affecting aerosol loads and their dependence on temperature over the studied region. Our results showed that the level of AOD in the southeastern US is mainly governed by anthropogenic emissions but the observed temperature dependent behaviour is most likely originating from non-anthropogenic emissions. Model simulations indicated that biogenic emissions of volatile organic compounds (BVOC) can explain the observed temperature dependence of AOD. According to the remote sensing data sets, the non-anthropogenic contribution increases AOD by approximately 0.009±0.018 1/K while the modelled BVOC emissions increase AOD by 0.022±0.002 1/K. Consequently, the regional direct radiative effect (DRE) of the non-anthropogenic AOD is -0.43±0.88 W/m2/K and -0.17±0.35 W/m2/K for clear- and all-sky conditions, respectively. The model estimate of the regional clear-sky DRE for biogenic aerosols is also in the same range: -0.86±0.06 W/m2/K. These DRE values indicate significantly larger cooling than the values reported for other forested regions. Furthermore, the model simulations showed that biogenic emissions increased the number of biogenic aerosols with radius larger than 100 nm (N100, proxy for cloud condensation nuclei) by 28 % per one degree temperature increase. For the total N100, the corresponding increase was 4 % which implies that biogenic emissions could also have a small effect on indirect radiative forcing in this region.

### Quantifying and Attributing Changes in Tropospheric Ozone over the Past Decade: The View from Aura

Jessica L. Neu (JPL; jessica.l.neu@jpl.nasa.gov), A. Sasha Glanville, Doug Kinnison, Hilke Oetjen, Willem Verstraeten, Nickolay Krotkov, Lok Lamsal, Nathaniel Livesey, Gloria Manney, Michelle Santee, Kevin Bowman, John Worden

Chemistry-climate models predict an intensification of the stratospheric circulation over the next century in response to greenhouse gas forcing, with uncertain consequences for tropospheric ozone and climate. We examine the role of the stratosphere in present-day tropospheric ozone variability and trends as measured by the Tropospheric Emission Spectrometer (TES) and Microwave Limb Sounder (MLS) onboard NASA's Aura satellite. Our previous work has shown that interannual variability in the stratospheric circulation of  $\pm 40\%$  leads to changes of  $\pm 25\%$  in northern midlatitude lower stratospheric ozone and  $\pm 2\%$  in northern midlatitude tropospheric ozone. Here we examine in more detail the drivers of stratospheric circulation variability and the subsequent ozone response using the Whole Atmosphere Chemistry-Climate Model (WACCM) in order to better understand the relationship between interannual and long-term changes in circulation and ozone. We find that both the stratospheric Quasi-Biennial Oscillation (QBO) and El Nino / Southern Oscillation (ENSO) drive stratospheric circulation changes but that both the circulation changes and ozone response depend critically on the timing of OBO and ENSO relative to one another and to the seasonal cycle. We also examine the role of stratospheric variability and changing emissions in tropospheric ozone trends over the past decade, with a focus on Eastern China and the Western United States. We discuss the implications of our work in terms of reducing uncertainties in long-term projections of tropospheric ozone.

#### Peroxyacetyl Nitrate (PAN) from TES: A new view into ozone chemistry

# Vivienne Payne (JPL/Caltech; vivienne.h.payne@jpl.nasa.gov), Emily Fischer, Zhe Jiang, Liye (Juliet) Zhu, Susan Kulawik, John Worden and Karen Cady-Pereira

Peroxyacetyl nitrate (PAN) is a thermally unstable reservoir for NOx that allows NOx to be transported large distances, enabling ozone formation far downwind from the original source. Sources of PAN precursors include anthropogenic combustion, biomass burning, lightning and biogenic emissions. PAN chemistry plays a key role in determining the global ozone distribution. TES PAN products are included in the TES v7 Level 2 data release. We show TES PAN associated with different sources. We show examples where elevated observations of PAN are clearly influenced by boreal fires. We also show that that TES observes enhanced PAN downwind of convective events over the US in summertime, suggesting potential for TES measurements to aid in placing constraints on the influence of lightning NOx on ozone production. PAN is validated with comparisons to aircraft measurements from the FRAPPE (2014) and ARCTAS (2008) campaigns and with the GEOS-Chem global chemical transport model. In addition, we present initial analysis of PAN from recent TES special observations over megacities, and discuss implications for export of pollution from these cities.

### Estimates of Lightning NOx Production from OMI NO2 Observations: A Pilot Study over the Gulf of Mexico and a Mid-latitude Continental Analysis

## Kenneth Pickering (NASA/GSFC; Kenneth.E.Pickering@nasa.gov), Dale Allen, Eric Bucsela, Allison Ring, Robert Holzworth, Nickolay Krotkov

We first evaluate nitrogen oxide (NOX= NO + NO2) production from lightning over the Gulf of Mexico region in a pilot study using data from the Ozone Monitoring Instrument (OMI) aboard NASA's Aura satellite along with detection efficiency-adjusted lightning data from the World Wide Lightning Location Network (WWLLN). A special algorithm was developed to retrieve the lightning NOx (LNOx) signal from OMI. The algorithm in its general form takes the total slant column NO2 from OMI and removes the stratospheric contribution and includes an air mass factor appropriate for the profile of lightning NOx to convert the slant column LNO2 to a vertical column of LNOx. WWLLN flashes are totaled over a period of 3 hours prior to OMI overpass, which is the time an air parcel is expected to remain in a 10 x 10 grid box. The analysis is conducted for grid cells containing flash counts greater than a threshold value of 3000 flashes that yields an expected LNOx signal greater than the background. Pixels with cloud radiance fraction (CRF) greater than a criterion value (0.9) indicative of highly reflective clouds are used. Results for the summer seasons during 2007 – 2011 yield mean LNOx production of  $\sim 80 \pm 45$  moles per flash over the region for regression and summation methods after accounting for biases and uncertainties. A different approach was used to estimate LNOx production for the same period of time over the northern mid-latitude continental regions of North America, Europe and East Asia. In this analysis a stratospheric and tropospheric background is subtracted, and the resultant LNOx is summed over the grid cells meeting the CRF criterion in the regions and months considered. This sum is divided by the sum of the associated detection efficiencyadjusted WWLLN flashes in the two hour period prior to OMI overpass. We obtain results (mean LNOx production of  $80 \pm 50$  moles per flash) consistent with the Gulf of Mexico study. Applying a regression approach to the mid-latitude data indicates that a nonlinear relationship may exist between flash rate and LNOx production. Smaller production per flash appears to be associated with greater flash rates. Our results are near the low end of literature estimates of LNOx production per flash but more robust than many prior estimates due to the large number of storms considered. The results are sensitive to several substantial sources of uncertainty.

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### Quick Look at the NASA ATom mission profiling and model-measurement analysis

Michael J. Prather (UC Irvine; mprather@uci.edu)

In late August 2016, the NASA ATom (Atmospheric Tomography) atmospheric chemistry mission will have completed its first 10-flight seasonal deployment, with the 2nd of 4 deployments scheduled for February 2017. All the measurements are still preliminary and not in final form. In this report, we look at some examples of the profiling capability of the DC-8 (0.5 to 12 km), which, safety permitting, will be done continuously from the Arctic to the edge of Antarctica. ATom should be able to help satellite retrievals of tropospheric chemical composition by measuring the covariance of key chemical constituents for profiles across the remote Pacific and Atlantic Ocean basins. This talk will also present a pre-ATom look at the model-measurement comparisons to be delivered as part of the ATom mission. Here we examine the reactivity of air parcels as sampled in six global chemistry models. The ATom analysis will examine the chemical climatology of regions weighted by their importance for the chemical evolution of methane and ozone.

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#### A method to account for surface BRDF effects on satellite UV/VIS algorithms

Wenhan Qin (SSAI; wenhan.qin@ssaihq.com), A. Vasilkov, N. Krotkov, L. Lamsal, R. Spurr, D. Haffner, J. Joiner

Most satellite nadir ultraviolet and visible cloud, aerosol and trace gaps algorithms make use of climatological surface reflectivity databases. Current LER (Lambertian Equivalent Reflectivity) product used in OMI NO2 cloud retrieval algorithms is based on monthly gridded LER climatologies that have no dependence on observation geometry. However, LER over land or ocean is sensitive to the sun-sensor geometry because surface is non-Lambertian and non-homogeneous; different pixels have different sun-view geometries and size of footprint. In this presentation, we propose a new concept – geometrydependent LER to account for surface BRDF and pixel size effects. The high resolution MODIS BRDF product and land-water map are used to compute OMI pixel-averaged BRDF coefficients and land fraction for land. Over ocean, Giss Cox-Munk model is used to model the glint reflection and Case 1 water model to calculate water leaving radiance contribution. A vector atmospheric radiative transfer model (Vlidort) is used to calculate LER components and TOA radiance over non-Lambertian surfaces. The geometrydependent LER is then computed on a satellite pixel basis. Its applications to current OMI NO2 and cloud algorithms as well as future satellite missions are finally discussed.

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## Composition of the Asian summer monsoon anticyclone: Climatology and variability from 10 years of Aura Microwave Limb Sounder measurements

Michelle L. Santee (JPL; Michelle.L.Santee@jpl.nasa.gov), G.L. Manney, N.J. Livesey, J.L. Neu, M.J. Schwartz, W.G. Read

Satellite measurements are invaluable for investigating the composition of the upper troposphere / lower stratosphere (UTLS) in the region of the Asian summer monsoon (ASM) anticyclone, which has been sparsely sampled by other means. The Aura Microwave Limb Sounder (MLS) makes simultaneous co-located measurements of trace gases and cloud ice water content (IWC, a proxy for deep convection) in the UTLS on a daily basis. Here we exploit the dense spatial and temporal coverage, long-term data record, and extensive measurement suite of Aura MLS to characterize the climatological composition of the ASM anticyclone and quantify its considerable spatial, seasonal, and interannual variability. We relate the observed trace gas behavior to various meteorological quantities, such as the size and strength of the ASM anticyclone, the extent and intensity of deep convection, and variations in the tropopause and the upper tropospheric jets in that region. Multiple species of both tropospheric and stratospheric origin are examined to help assess whether the observed variability arises from variations in transport processes or changes in the strength or location of surface emissions.

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#### Comparisons of ACE-FTS and MLS v4.2 atmospheric profiles and drift analysis

Patrick E. Sheese (University of Toronto; psheese@atmosp.physics.utoronto.ca), Kaley A. Walker, Chris D. Boone, Jason Zou

The ACE-FTS (Atmospheric Chemistry Experiment – Fourier Transform Spectrometer) instrument on board the Canadian SCISAT-1 satellite has been measuring profiles of atmospheric temperature and concentrations of 30+ trace gases since February 2004. Many of these trace gases are also measured by the MLS (Microwave Limb Sounder) instrument on board the Aura satellite, namely O3, H2O, N2O, HNO3, CO, HCl, HCN, and CH3Cl. This study will investigate the systematic differences between the correlative ACE-FTS and MLS data sets of these species and temperature. How the systematic differences are changing with time will also be investigated, with a focus on O3, H2O, and HCl. For each species, at each altitude level, drift analysis will make use of segmented linear regression in order to identify any breakpoints in the relative difference time series.

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### In-flight performance of the Ozone Monitoring Instrument

V. M. Erik Schenkeveld (KNMI; Erik.Schenkeveld@knmi.nl), G. Jaross, S. Marchenko, D. Haffner

In this presentation results of in-flight performance analysis of OMI will be presented. Performance and calibration data of the 12-year mission has been analyzed. Results of detector degradation, radiometric calibration, spectral calibration and row anomaly evolution will be shown. The instrument degrades gracefully, with a degradation of 3-8 % in irradiance and 1-2 % in radiances. The wavelength registration is stable to 0.005 - 0.020 nm.

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# The Impact of Gravity Waves and Cloud Nucleation Threshold on Stratospheric Water and Tropical Tropospheric Cloud Fraction

Mark Schoeberl (STC; mark.schoeberl@mac.com), Andrew Dessler, Hao Ye, Tao Wang, Melody Avery, Eric Jensen

Using the Modern Era Retrospective-Analysis for Research and Applications (MERRA) and MERRA-2 reanalysis winds, temperatures and anvil cloud ice, we explore the impact of varying the cloud nucleation threshold relative humidity (RH) and high frequency gravity waves on stratospheric water vapor (H2O) and upper tropical tropopause cloud fraction (TCF). Our model results are compared to 2008/9 winter TCF derived from Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and H2O observations from the Microwave Limb Sounder (MLS). The RH threshold affects both model H2O and TCF while high frequency gravity waves mostly impact TCF. Adjusting the nucleation RH and the amplitude of high frequency gravity waves allows us to tune the model to observations. Reasonable observational agreement is obtained with a nucleation threshold between 130% - 150% RH consistent with airborne observations. For the MERRA reanalysis, we lower the tropopause temperature by 0.5 K roughly consistent with GPS radio occultation measurements and include ~0.1 K high frequency gravity wave temperature oscillations in order to match TCF and H2O observations. For MERRA-2 we do not need to adjust the tropopause temperature nor add gravity waves, because there are sufficient high frequency temperature oscillations already present in the MERRA-2 reanalysis to reproduce the observed TCF.

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### Interannual Variability in MLS Trace Gas Observations in the Antarctic Polar Vortex: Issues in Detection and Attribution of Trends

Michael J. Schwartz (JPL/Cal Tech; michael.j.schwartz@jpl.nasa.gov), Michelle L. Santee, Nathaniel J. Livesey, Gloria L. Manney

The Microwave Limb Sounder (MLS), launched on NASA's Aura satellite in 2004, measures an extensive suite of atmospheric constituents involved in stratospheric ozone chemistry, including major chlorine and nitrogen reservoir and radical species, long-lived tracers, and ozone itself. We use the 12-year Aura MLS record in conjunction with meteorological analyses to quantify interannual variability and longer-term changes in lower stratospheric trace gas mixing ratio histograms and relate them to dynamical and meteorological variations. We focus on the Antarctic, where unambiguous trends in constituent behavior should be more readily detected than in the more highly dynamically variable Arctic. Even in the Antarctic, however, meteorological variability induces substantial year-to-year variations in trace gas distributions. Although decreases in lower stratospheric ozone loss can be expected in response to declining abundances of ozonedepleting substances, many confounding factors complicate attribution of apparent longer-term changes in ozone and other species to particular processes. Analysis of meteorological fields (including calculated mixing diagnostics) will be combined with histograms of long-lived tracers to provide insight into the possible significance of changes in dynamical and transport processes, including polar vortex characteristics and the location and strength of its "boundary" as well as potential changes in the large-scale transport circulation. This approach will be used to assess the degree to which chemical and dynamical processes controlling the evolution of ozone, as reflected in the frequency distributions of its abundance in the lower stratosphere, can be distinguished from one another.

# Upper Tropospheric Water Vapor Simulated in CMIP5 Models: From Present-day Variations to Long-term Change under Global Warming

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#### Hui Su (JPL; Hui.Su@jpl.nasa.gov), Hanii Takahashi, Jonathan H. Jiang

Upper tropospheric water vapor (UTWV) plays a critical role in amplifying global warming caused by increasing greenhouse gases, yet it is one of the most poorly simulated quantities in climate models. We diagnose the UTWV simulation errors from CMIP5 models by using Aura and other "A-Train" satellite observations and reanalysis data. It is found that models generally have positive biases in UTWV, except over the continental convective regions where negative biases predominate. The fractional UTWV errors show large positive errors over the large-scale descending regimes ( $0 < \omega 500 < 40$  hPa/day) where large model spreads also exist. The errors for UTWV interannual anomalies are abundant over the climatologically deep convective regions (SST > 300 K or  $\omega 500 < -30$  hPa/day) and these errors are positive (negative) where anomalous descent (ascent) occurs during El Niño. We find that the water vapor errors are dominated by the

errors in relative humidity (RH) rather than in temperature throughout the troposphere, while temperature errors play an important role for water vapor errors near the tropopause. Under the RCP4.5 global warming scenario, the simulated mean fractional UTWV changes between 300 and 100 hPa range from 12.4 to 28.0 %/K across all models while the fractional water vapor changes are about 5–8 %/K in other regions and at lower altitudes. The "upper-tropospheric amplification" of the water vapor change is primarily driven by a larger temperature increase in the upper troposphere than in the lower troposphere per degree of surface warming. The relative contributions of atmospheric temperature and RH changes to the water vapor change in each model vary between 71.5 to 131.8 % and 24.8 to -20.1 %, respectively. The inter-model differences in the water vapor change is primarily caused by differences in temperature change, except over the inter-tropical convergence zone within 10°S-10°N where the model differences due to the RH change are significant. Furthermore, we find that there is generally a positive correlation between the rates of water vapor change for long-term surface warming and those on the interannual time scales. However, the rates of water vapor change under long-term warming have a systematic offset from those on the inter-annual time scales and the dominant contributor to the differences also differs for the two time scales, suggesting caution needs to be taken when inferring long-term water vapor changes from the observed interannual variations

# What solar measurements can tell about instrument transfer function—lessons learned from OMI and OCO-2

### Kang Sun (Harvard-Smithsonian Center for Astrophysics; kang.sun@cfa.harvard.edu), Xiong Liu, Gonzalo González Abad, Zhaonan Cai, Caroline Nowlan, Kai Yang, Kelly Chance

The accurate characterization of instrument transfer function (ITF) is essential for the spectral calibration of space-borne grating spectrometers and the retrieval of the Earth's atmospheric constituents. For instruments using 2-D detectors like OMI and OCO-2, the ITF are complicated functions of both wavelength (the column dimension) and crosstrack viewing direction (the row dimension). Preflight measurements, parameterized as hybrid Gaussian functions (OMI) or saved in tabulated forms (OCO-2), are often used as constant input in spectral calibration and trace gas retrieval. However, the launching process and the thermal/radiative contrast between the space and laboratory conditions may induce subtle changes in the ITF. The on-orbit thermal variation, instrument degradation, and switching of observation modes may also cause ITF variations. This study utilizes the frequent solar measurements from OMI/OCO-2 and the recent development of high-resolution, high-accuracy reference solar spectrum to characterize the on-orbit behavior of their ITF. The ITF are fitted using various forms of analytical functions or variants of the preflight ITF. The cross-track dependence of OMI ITF is significantly different from the preflight measurements. The ITF full width at half maximum (FWHM) of detector pixels at large viewing angles are up to 10% higher than the preflight ITF FWHM. Nonetheless, the OMI ITF is stable over time (10 years of

operation), despite of the decaying of detector SNR and the occurrence of OMI row anomaly. Because OCO-2 targets much higher retrieval precision and spectral resolution, its ITF cannot be adequately parameterized by analytical functions. A "stretch" and a "sharpen" term are used instead to capture the ITF shape change. Widening of the wings of OCO-2 ITF by 2% was found between decontamination events, which is independently verified by the solar induced fluorescence (SIF) retrieval using the earthshine spectra. These results have implications for future satellites targeting both short-lived species in the UV/visible and long-lived species in the infrared. The on-orbit behavior of ITF needs to be carefully investigated for accurate retrieval.

# Using OMI NO2 as top-down constraint on US NOx emissions: complication from the upper troposphere

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### Katherine Travis and <u>Daniel Jacob (Harvard; djacob@fas.harvard.edu; presenting)</u>, Ronald Cohen (UC Berkeley), Thomas Ryerson (NOAA)

Aircraft observations of NOx and its oxidation products during the NASA SEAC4RS and NOAA SENEX campaigns over the Southeast US in summer 2013 suggest that anthropogenic NOx emissions in the US EPA National Emission Inventory (NEI) are too high by a factor of two. This is supported by national network observations of nitrate wet deposition fluxes. OMI NO2 observations from both the NASA (operational) and UC Berkeley (BEHR) products are consistent with such a large NEI overestimate of NOx emissions but interpretation of the OMI data is complicated by a large contribution of the upper troposphere to total tropospheric NO2 columns. Median aircraft vertical profiles during SEAC4RS indicate comparable NO2 mixing ratios in the upper and lower troposphere, such that the upper troposphere contributes 30-50% of the total NO2 tropospheric column sensed by OMI. Such a large contribution of the upper troposphere reflects the relatively low US NOx emissions (a consequence of strong emission controls over the past decade) and a large source of lightning NOx. Observations of the NO2/NO ratio in the upper troposphere further indicate values larger than expected from NO-NO2-O3 photochemical equilibrium, and explainable in part by convective injection of peroxy radical precursors (such as formaldehyde and methyl hydroperoxide) to the upper troposphere. Better understanding of the factors controlling upper tropospheric NO2 is critical for using satellite observations of NO2 columns as top-down constraints on NOx emissions.

# **Resolving Model-Observation Discrepancy in the Mesospheric and Stratospheric HOx Chemistry**

# Shuhui Wang (UCLA; shuhui@ucla.edu), Qiong Zhang, King-Fai Li, Stanley Sander, Yuk Yung

The variability in middle atmospheric HOx, mainly hydroxyl (OH) and hydroperoxyl (HO2), plays a key role in controlling ozone. Earlier studies about solar cycle induced OH variability show surprisingly large discrepancies between model and observations. Uncertainties in chemical kinetics as well as the solar irradiance variability could contribute to these discrepancies. In this study, we examine the uncertainties in middle atmospheric HOx chemistry by comparing the Aura Microwave Limb Sound (MLS) OH and HO2 measurements with the simulations of the Caltech-JPL KINETICS photochemical model. The model using the standard chemical kinetics underestimates OH and HO2 concentrations in the mesosphere. To resolve the discrepancies, we use MLS OH and HO2 measurements as benchmark to adjust the involved chemical rate coefficients within reasonable uncertainty ranges with an optimal estimation algorithm. The results show that four key reaction rate constants and the O2 cross section at Lyman- $\alpha$  (121.6 nm) need to be modified to achieve optimal model results that match observations. Model simulation with the adjusted parameters shows significantly better agreements with both OH and HO2 measurements. In particular, the rate coefficient of H  $+ O2 + M \rightarrow HO2 + M$  requires a very large adjustment beyond the recommended uncertainty, which calls for future laboratory measurements. An alternative is a radiative association reaction:  $H + O2 \rightarrow HO2 + hv$ , which has never been measured. Our study indicates that accurate photochemical rate coefficients and molecular cross sections are crucial in simulating stratospheric and mesospheric HOxchemistry. High quality satellite observations can be used to constrain or retrieve photochemical parameters and help improve our understanding of atmospheric chemistry.

#### Effectiveness of air quality policy for SO2 and NOx emissions in China

Ronald J. van der A (KNMI; avander@knmi.nl), Bas Mijling, Jieying Ding, Maria Elissavet Koukouli, Fei Liu, Qing Li, Huiqin Mao, Nicolas Theys

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To study air quality trends in China for the period 2005-2015 we derive SO2 columns and NOx emissions on a provincial level from satellite observations of OMI. To put these trends into perspective they are compared with public data on energy consumption and the environmental policies of China. We distinguish the effect of air quality regulations from economic growth by comparing them relatively to fossil fuel consumption. Pollutant levels, per unit of fossil fuel, are used to assess the effectiveness of air quality regulations. Despite an increasing fossil fuel consumption and a growing transport sector, the effects of air quality policy in China are clearly visible. Especially the last three years show a strong reduction in SO2 and NOx emissions per fossil fuel unit, since the authorities have implemented several new environmental regulations. Without the air quality regulations the concentration of SO2 would be almost 3 times higher and the NO2 concentrations would be at least 30% higher than they are today in China.

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#### Effects of surface BRDF on OMI cloud and NO2 retrievals

Alexander Vasilkov (SSAI; alexander.vasilkov@ssaihq.com), W. Qin, N. Krotkov, L. Lamsal, R. Spurr, D. Haffner, J. Joiner, E.-S. Yang, S. Marchenko

To account for the surface BRDF effects in OMI cloud and NO2 retrievals we have developed a novel method based on a concept of geometry-dependent surface Lambertian-equivalent reflectivity (LER). The geometry-dependent LER is calculated using a vector radiative transfer model (VLIDORT) with high spatial resolution BRDF information from MODIS over land and the Cox-Munk slope distribution over ocean, coastal and in land water accounting for contribution from water-leaving radiance. Implementation within the existing OMI cloud and NO2 retrieval framework requires replacement of the climatological surface reflectivity databases used in the cloud and NO2 algorithms. A detailed comparison of the effective cloud fractions and pressures derived with climatological and geometry-dependent LERs is carried out. The effects on retrieved cloud parameters are relatively small on average and diminish with increasing cloud fraction. Even though the impact is small on average, for individual FOVs it can be as large as  $\pm 0.05$  for the cloud fraction and 100 hPa for the cloud pressure. The BRDF effects are noticeably larger for the O2-O2 cloud algorithm that uses visible wavelengths as compared with the rotational Raman scattering cloud algorithm that utilizes a UV spectral range. Geometry-dependent LER and corresponding retrieved cloud products are then used as inputs to OMI standard NO2 algorithm. We find that the use of geometrydependent LER increases the NO2 vertical columns by up to 50% over highly polluted areas. Only minor changes to NO2 columns (within 5%) are found over unpolluted and cloudy areas.

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#### **TROPOMI on Sentinel 5 Precursor: Ready for Launch**

J. Pepijn Veefkind (KNMI / TU Delft; veefkind@knmi.nl), Q. Kleipool, A. Ludewig, I. Aben, P.F. Levelt, TROPOMI team

The Copernicus Sentinel 5 Precursor (S5P) is the first of the Sentinels dedicated to monitoring of the atmospheric composition. The mission objectives of S5P are to monitor air quality, climate and the ozone layer, in the time period between 2016 and 2022. S5P will fly in a Sun-synchronized polar orbit with a 13:30 hr local equator crossing time. The single payload of the S5P mission is TROPOspheric Monitoring Instrument (TROPOMI), which is developed by The Netherlands in cooperation with ESA. TROPOMI is a nadir viewing shortwave spectrometer that measures in the UV-visible wavelength range (270-

500 nm), the near infrared (710-770 nm) and the shortwave infrared (2314-2382 nm). TROPOMI will have an unprecedented spatial resolution of 7x7 km2 at nadir. The spatial resolution is combined with a wide swath to allow for daily global coverage. The TROPOMI/S5P geophysical (Level 2) operational data products include nitrogen dioxide, carbon monoxide, ozone (total column, tropospheric column profile), methane, sulphur dioxide, formaldehyde and aerosol and cloud parameters. The main heritage for TROPOMI comes from the Ozone Monitoring Instrument (OMI) for the measurement principle, and from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) for the shortwave infrared band. Many of the lessons learned in these missions have resulted in design improvements for TROPOMI. One of the most striking features of TROPOMI is the high spatial resolution of 7x7 km2 at nadir. The high spatial resolution serves two goals: (1) emissions sources can be detected with a higher accuracy and (2) the number of cloud-free ground pixels will increase substantially. The higher spatial resolution is also combined with a significantly higher signal-to-noise ratio per ground pixel, compared to OMI. This will further enhance the capabilities of TROPOMI to detect small emissions sources. The S5P will fly in a socalled loose formation with the U.S. Suomi NPP (National Polar-orbiting Partnership) satellite. The primary objective for this formation flying is to use the cloud clearing capabilities of the VIIRS (Visible Infrared Imager Radiometer Suite). The temporal separation between TROPOMI and VIIRS will be less than 5 minutes. Once this formation has been established, it will enable synergistic data products and scientific research potentials. In this contribution we will present the TROPOMI mission, with a focus on the performance and the operational data products.

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#### NOX emissions over European cities revisited: a top-down approach

Willem Verstraeten (Royal Meteorological Institute of Belgium; willem.verstraeten@meteo.be), Boersma, K.F., Ndouros, J., Williams, J.E., Eskens, H., Delcloo, A.

High levels of nitrogen oxides, (NOX = NO + NO2), adversely impact the human and ecosystem health at the surface and play a key role in tropospheric chemistry. NOX emissions drive major processes in regional and global chemistry transport models (CTM). Although accurate inventories are essential, state-of the- art emission databases may vary substantially and uncertainties are high since reported emissions factors may differ in order of magnitude and more. NOX is mainly generated in polluted regions by anthropogenic combustion from industrial, traffic and household activities typically observed in large and densely populated urban areas. Satellite sensors have demonstrated excellent abilities to capture tropospheric NO2 columns over many large cities worldwide. Here we revisit NOX emissions over 21 selected European urban areas using the exponentially modified Gaussian (EMG) method and the effect of wind on tropospheric NO2 distributions from different datasets to estimate the effective NO2 life time and the average summertime (April-September 2013) NOX emissions. OMI (Ozone Monitoring Instrument) tropospheric NO2 column data and datasets derived from the Chemical Transport Model LOTOS-EUROS are used, including a LOTOS-EUROS data set assimilating OMI observations. LOTOS-EUROS is run with MACC NOX emissions inputs. Comparison is made with surface measurements of NO2 where available. First results indicate that the NOX derived from the different datasets are in general in the same order of magnitude when the general error ranges that apply on these data sets are considered.

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# **Rapid increases in tropospheric ozone production and export from China:** A view from AURA

Willem Verstraeten (Royal Meteorological Institute of Belgium (KMI); willem.verstraeten@meteo.be), Neu J.L., Williams J.E., Bowman K.W., Worden J.R., Boersma K.F.

Eastern Asia has the fastest growing anthropogenic emissions in the world, possibly affecting both the pollution in the local troposphere as well as in the trans-Pacific region. Local measurements over Asia show an increase of tropospheric ozone (O3), which might contribute to the observed positive O3 trends over western US. To date, there was no unambiguous evidence showing that enhanced Asian pollution is responsible for these trends. Here we show that large air pollution import from China offsets the local efforts to improve air quality with respect to O3 in the 3-9 km partial column over the western US with more than 40%. These results are based on observations of tropospheric O3 from TES (Tropospheric Emission Spectrometer, onboard AURA), tropospheric NO2 measurements from OMI (Ozone Monitoring Instrument, onboard AURA) and lower stratospheric observations of O3 from MLS (Microwave Limb Sounder, onboard AURA). These satellite observations are combined with model runs of the TM5 chemistry transport model. TM5 is able to reproduce the 2005-2010 observed rapid rise in free tropospheric O3 of 7% over China from TES once OMI NO2 measurements were implemented in TM5 to update NOX emissions. MLS observations on lower stratospheric O3 have the potential to improve the stratosphere-troposphere exchange (STE) estimate in TM5 which is mainly driven by ECMWF meteorological fields. Constraining the TM5 modeled trend of the STE contribution to the 3-9 km partial O3 column using MLS observations of stratospheric O3 lead to a better explanation of the sources of the free tropospheric O3 trends over China. Based on the OMI inferred TM5 updates in NOX emissions, the impact of Asian O3 and its precursors on the free troposphere (3-9 km) over the western US could be quantified using TM5. Long-range transport of pollution emphasis the need for global effort to address both the global as well as the regional air quality and climate change.

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# **Recent Results from and Mission Status of the Atmospheric Chemistry Experiment** (ACE)

# Kaley A. Walker (University of Toronto; kaley.walker@utoronto.ca), Patrick E. Sheese, Peter F. Bernath, Chris Boone, and C. Thomas McElroy

In August 2016, the Canadian-led Atmospheric Chemistry Experiment (ACE) mission will complete its thirteenth year in orbit on board the SCISAT satellite. SCISAT/ACE uses infrared and UV-visible spectroscopy to investigate the chemistry and dynamics of the Earth's atmosphere. The long lifetime of ACE has provided a valuable time series of composition measurements that contribute to our understanding of ozone recovery, climate change and pollutant transport. The primary instrument on board, the ACE Fourier Transform Spectrometer (ACE-FTS) is a high-resolution (0.02 cm-1) infrared FTS operating between 750 and 4400 cm-1. It also contains two filtered imagers (0.525 and 1.02 microns) to measure atmospheric extinction by clouds and aerosols. The second instrument is a dual UV-visible-NIR spectrophotometer called ACE-MAESTRO (Measurements of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation) which was designed to extend the ACE wavelength coverage to the 280-1030 nm spectral region. From these solar occultation measurements, altitude profiles of atmospheric trace gas species, temperature and pressure are obtained. In addition to the mission and instrument status, a review of current science results from the ACE mission will be presented.

### Validation of OMI and GOME-2A and GOME-2B tropospheric NO2, SO2 and HCHO products using MAX-DOAS observations in Wuxi, China: effects of coincidence criteria, clouds and a-priori profiles

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Tropospheric vertical column densities (VCDs) of NO2, SO2 and HCHO derived from the Ozone Monitoring Instrument (OMI) on AURA and Global Ozone Monitoring Experiment 2 aboard METOP-A (GOME-2A) and METOP-B (GOME-2B) are widely used to characterize the global distributions, trends, dominating sources of the trace gases and for the comparison with chemical transport models (CTM). We use tropospheric VCDs and vertical profiles of NO2, SO2 and HCHO derived from MAX-DOAS measurements from 2011 to 2014 in Wuxi, China, to validate the corresponding products derived from OMI and GOME-2A/B by different scientific teams (daily and bimonthly averaged data). Prior the comparison we investigate the effects of the spatial and temporal coincidence criteria for MAX-DOAS and satellite data on the comparison results. We find that the distance of satellite data from the location of the MAX-DOAS station is the dominating effect, and we make suggestions for the spatial and temporal averaging. We also investigate the effect of clouds on both MAX-DOAS and satellite observations. Our results indicate that the discrepancies between satellite and MAX-DOAS results increase with increasing effective cloud fraction and are dominated by the cloud effect on the satellite products. We find a systematic underestimation of all SO2 and HCHO products and an overestimation of the GOME-2A/B NO2 products (the DOMINO NO2 product is only slightly underestimated). To better understand the reasons for the differences, we recalculated the AMFs for satellite observations based on the shape factors (SFs) derived from MAX-DOAS. The recalculated satellite VCDs agree better with the MAX-DOAS VCDs than those from the original products, especially for large VCDs. Finally we find that the satellites systematically overestimate the magnitude of the diurnal variations of NO2 and HCHO. No significant weekly cycle for all trace gases is found by either the satellites or the MAX-DOAS measurements.

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#### EOS Aura data in GMAO's MERRA-2 reanalysis

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Modern-Era Retrospective Analysis for Research and Applications 2 (MERRA-2) is a new global atmospheric reanalysis, covering the time period from 1980 to present, currently produced by NASA/GMAO. MERRA-2 is generated with a recent version of the GEOS-5 data assimilation system that includes numerous modifications to the general circulation model, data assimilation module and the observing system, relative to the previous reanalysis, MERRA. In particular, starting in 2004 MERRA-2 assimilates stratospheric ozone and upper stratospheric temperature profiles from the Microwave Limb Sounder (MLS) as well as total ozone data from the Ozone Monitoring Instrument (OMI) flying on the EOS Aura satellite. In this presentation we will highlight some validation results of the new reanalysis focusing on its suitability for scientific applications. Specifically, we will discuss the impacts that the Aura data have on the quality of the reanalysis ozone and upper stratospheric temperature. Consistent with our previous results, assimilation of MLS data leads to a very good representation of the spatial and temporal variability of the stratospheric ozone in MERRA-2, including the tropopause region, as illustrated by comparisons against independent observations from ozonesondes, MIPAS and SAGE II. For example, the agreement between the MERRA-2 ozone and MIPAS in terms of the difference standard deviation in the lower stratosphere improves by a factor of 1.6 in the Aura period compared to the MERRA reanalysis. MLS temperature profiles are assimilated at pressures higher than 5 hPa from August 2004 onwards leading to a more realistic representation of the stratopause in MERRA-2 compared to MERRA and the pre-Aura period in MERRA-2. This will be illustrated by examples of the stratopause reformation following sudden stratospheric warming events and the temporal continuity of upper stratospheric temperature during the Aura period in MERRA-2.

### What's NEW at the GES DISC: Evolution of data management and services for Aura mission and beyond

Jennifer Wei (NASA GES DISC/Adnet; jennifer.c.wei@nasa.gov), James Johnson, Irina Gerasimov, Bruce Vollmer, Steve Kempler

NASA GES DISC strives to provide end users with nearly immediate access and interactivity across GES DISC data. Over recent years, GES DISC has been evolving and improving our data management and services in order to promote GES DISC data to be easily discovered, navigated, and interoperated. As a result, we will present a brief review of new and upcoming data services at the GES DISC including the Unified User Interface (UUI), Common Metadata Repository (CMR), Subsetting services in Satellite Level 1/2 datasets, Data Lists, User Forum, Interactive Visualization and Analysis online tool (Giovanni), and Satellite Level 2 visualizing image capabilities. We are soliciting suggestions regarding the user interface and additional Data List use cases for continuing serving Aura and Sentinel 5P communities.

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### TES participation in the IGAC/TOAR (Tropospheric Ozone Assessment Report)

Helen Worden (NCAR; NCAR; hmw@ucar.edu), Jessica Neu, Susan Kulawik, Catherine Wespes, Kevin Bowman

The mission of the IGAC/TOAR (Tropospheric Ozone Assessment Report) is to provide the research community with an up-to-date scientific assessment of tropospheric ozone's global distribution and trends from the surface to the tropopause, with global metrics for climate change, human health and crop/ecosystem research. The goals are to produce peer-reviewed chapters that will be combined into the first assessment report on tropospheric ozone and provide accessible, documented data on global ozone exposure and dose metrics for urban and non-urban sites. The TES and IASI teams are contributing climatologies, time series, and associated descriptions for ozone in the free troposphere. We will present some of the TES and IASI results that will be included in the TOAR and discuss comparisons to other free tropospheric ozone records.

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#### **Contribution of Fires to the Global Methane Budget**

John Worden (JPL; john.r.worden@jpl.nasa.gov), Anthony Bloom, Zhe Jiang, Helen Worden

Fire emissions of methane potentially represent a significant but highly uncertain component of the global methane budget with estimates ranging from 14 to 88 Tg / yr. This range of estimates is driven in part by knowledge of dry matter burned, the fuel type, and variations in combustion efficiency. Here we use global estimates of CO emissions

based on CO profiles from the NASA Terra MOPITT instrument and CH 4 /CO emission ratios from NASA Aura TES data to constrain estimates of the global contribution to atmospheric methane from fires. For the years 2001 to the present we find that fire emissions of methane are approximately 3% (or  $\sim 15$  Tg / yr) of the global methane budget. Furthermore, fire emissions have likely been decreasing during this time period indicating that non-fire methane fluxes have primarily contributed to the recent increase in global atmospheric methane concentrations.

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# Quantifying Lower Tropospheric Methane Concentrations Using GOSAT near-IR and TES thermal IR measurements

John R. Worden (JPL; john.worden@jpl.nasa.gov), Alex J. Turner, Anthony Bloom, Susan S. Kulawik, Robert Parker, Vivienne H. Payne

Evaluating surface fluxes of methane using total column data requires models to accurately account for the transport and chemistry of methane in the free troposphere and stratosphere, thus reducing sensitivity to the underlying fluxes. Vertical profiles of methane have increased sensitivity to surface fluxes because lower tropospheric methane is more sensitive to surface fluxes than a total column and quantifying free tropospheric CH4 concentrations helps to evaluate the impact of transport and chemistry uncertainties on estimated surface fluxes. Here we demonstrate new estimates of lower tropospheric CH4 concentrations through the combination of free tropospheric methane measurements from the Aura Tropospheric Emission Spectrometer (TES) and XCH4 (dry-mole air fraction of methane) from the Greenhouse Gases Observing Satellite Thermal And Near Infrared for Carbon Observations (GOSAT TANSO, herein GOSAT for brevity). The calculated precision of these estimates ranges from 10 to 30 ppb for a monthly average on a 4x5 latitude / longitude degree grid making these data suitable for evaluating lowertropospheric methane concentrations. Smoothing error is approximately 10 ppb or less. Comparisons between these data and the GEOS-Chem model demonstrate that these lower-tropospheric CH4 estimates can resolve enhanced concentrations over flux regions that are challenging to resolve with total column measurements. We also use the GEOS-Chem model and surface measurements in background regions across a range of latitudes to determine that these lower-tropospheric estimates are biased low by approximately 65 ppb, with an accuracy of approximately 6 ppb (after removal of the bias) and an actual precision of approximately 30 ppb. This 6 ppb accuracy is consistent with the accuracy of TES and GOSAT methane retrievals.

### Quality Assurance for NASA, KNMI, and QA4ECV spectral fitting algorithms for OMI NO2 Slant Columns

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Scientific parties from Europe and the United States challenge each other in developing accurate reliable NO2 slant column density (SCD) retrieval algorithms for the Ozone Monitoring Instrument (OMI). Both NASA and KNMI recently developed substantially revised spectral fitting algorithms for OMI NO2 SCD retrievals, named Goddard SCD [Marchenko et al., 2015], and OMNO2A v2.0 [van Geffen et al., 2015]. Within the framework of the EU FP7-project Quality Assurance for Essential Climate Variables (QA4ECV), a third 'community' algorithm has recently become available based on extensive comparisons of spectral fitting approaches between BIRA, the University of Bremen, MPI, and KNMI. These new algorithms differ in their approaches. The NASA Goddard algorithm is based on multiple small fitting windows optimized for iterative wavelength adjustments and Ring pattern removal. Then broad, customized windows are used for sequential retrieval of trace-gas species. The improvements in OMNO2A and QA4ECV focus on improving the level of physical detail in the fitting model within the wide 405-465 nm window. These new algorithms presumably constitute a major improvement over previous OMI NO2 fitting algorithms that resulted in SCDs that were biased high by 10-40%. Here we evaluate the new OMI NO2 SCD values and their uncertainties in detail. We compare the three different OMI NO2 data sets over the clean Pacific area. All three products have 20-30% lower values than the slant columns from OMNO2A v1.0, which was the point of departure for the widely-used Standard and DOMINO v2 products. Slant columns retrieved from the OA4ECV fitting algorithm agree within a few percent with the OMNO2A v2.0 SCDs. We move on and investigate the uncertainties in the slant column retrievals. SCD (spectral fitting) uncertainty arises from wavelength calibration, radiometric noise, cross-section uncertainty, and includes contributions from the "stripe" effect. We evaluate the uncertainty estimates provided by the fitting algorithms against independently determined statistical estimates of the product uncertainties over the clean Pacific. This unique check of the algorithm uncertainties is possible over this area which is dominated by almost invariable stratospheric NO2. Our preliminary results suggest that the SCD uncertainties provided in the various data products may be too pessimistic, especially for OMNO2A v1, where the fitting uncertainty exceeds the statistical uncertainty by most. For the OA4ECV and the OMNO2A v2 algorithms, there is much better agreement between the statistical and algorithm uncertainties. Moreover, the statistical uncertainty for the new QA4ECV SCDs is smaller than OMNO2A v2 and OMNO2A v1. This provides confidence in the improvements implemented in the QA4ECV spectral fitting approach. We will extend our comparison and also evaluate the lower uncertainties reported for the Goddard SCD relative to OMNO2A v1 against our clean-sector statistical estimate.

### Observing atmospheric formaldehyde from OMI: validation, intercomparison, trend analysis and public health implications

Lei Zhu (Harvard University; leizhu@fas.harvard.edu), Daniel J. Jacob, Patrick S. Kim, Jenny A. Fisher, Karen Yu, Katherine R. Travis, Loretta J. Mickley, Robert M. Yantosca, Melissa P. Sulprizio, Isabelle De Smedt, Gonzalo Gonzalez Abad, Kelly Chance, Can Li, Richard Ferrare, Alan Fried, Johnathan W. Hair, Thomas F. Hanisco, Dirk Richter, Amy Jo Scarino, James Walega, Petter Weibring, Glenn M. Wolfe

Formaldehyde (HCHO) column data from OMI are widely used as a proxy for emissions of volatile organic compounds (VOCs), but validation of the data has been extremely limited. Here we use highly accurate HCHO aircraft observations from the NASA SEAC4RS campaign over the Southeast US in August–September 2013 to validate the operational HCHO retrieval (OMI-SAO) and a research retrieval (OMI-BIRA). We also compare the two OMI retrievals with available retrievals from other satellites (GOME2A, GOME2B and OMPS) and from different groups. OMI retrievals feature a HCHO maximum over Arkansas and Louisiana, consistent with the aircraft observations and GEOS-Chem, and reflecting high emissions of biogenic isoprene. OMI retrievals are broadly consistent with other retrievals in spatial variability over the Southeast US  $(r=0.4-0.7 \text{ on a } 0.50 \times 0.50 \text{ grid})$  as well as day-to-day variability (r=0.5-0.8). Validation results show that OMI retrievals provide a reliable proxy for isoprene emission variability but with a low mean bias (-43% for OMI-SAO; -20% for OMI-BIRA) due both to the spectral fitting and the scattering weights used in the retrievals. We then apply the corrected OMI-SAO data to conduct the two following studies. First, we examine trend in HCHO columns from 2005 to 2014 over the North America. OMI clearly captures trends associated with anthropogenic emission control near Houston, oil/gas production increase over Oil Sands as well as land cover changes over the Southeast US. Second, we drive a fine surface ambient HCHO concentration map  $(0.20 \times 0.20)$  based on oversampled OMI HCHO columns, localized HCHO vertical profiles sampled from GEOS-Chem, and diurnal variations in surface HCHO measured at various sites. We estimate a total number of 7000 lifelong cancer risks due to exposure of surface ambient HCHO.

Land cover and seasonality effects on biomass burning emissions observed from satellites

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Trace gas emissions from biomass burning can vary greatly both regionally and from event to event, but our current scientific understanding is unable to fully explain this variability. The large uncertainty in ozone formation resulting from fire emissions has posed a great challenge for assessing fire impacts on air quality and atmospheric composition. Satellite observations from OMI offer a powerful tool to observe biomass burning events by providing observations globally over a range of environmental conditions that effect emissions of NOx, formaldehyde, and glyoxal. We have investigated the seasonal relationship of biomass burning enhancements of these trace gases derived from OMI observations over tropical South America, Africa, and Indonesia. Land cover type (also derived from satellite observations) has a significant impact on formaldehyde and glyoxal enhancements from fire activity. We have found that the chemical ratio between formaldehyde and glyoxal is dependent on the burned land type and will present our current hypotheses for the spatial variation of this ratio in the tropics. In individual case studies we will investigate how these chemical ratios can inform our knowledge of the secondary formation of VOCs and ozone.