ANALYSIS OF SATELLITE AND SUB-ORBITAL MEASUREMENTS

GRANT NNG04GE15G

Annual Report
For the period 1 March 2004 through 31 December 2004

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December 2004

Prepared for

National Aeronautics and Space Administration

Washington, DC 20546

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The Smithsonian Astrophysical Observatory
is a member of the
Harvard-Smithsonian Center for Astrophysics

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YEAR 1 PROGRESS REPORT
NASA Grant NNG04GE15G

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Performance period: 3/1/04-12/31/04

PREPARING FOR AND SUPPORTING INTEX THROUGH INTEGRATED ANALYSIS OF SATELLITE AND SUB-ORBITAL MEASUREMENTS WITH GLOBAL AND REGIONAL 3-D MODELS

1. Project Objective

The objective of this project is to support the INTEX aircraft mission by developing experience in the integrated analysis of existing sub-orbital observations and satellite observations with numerical models. Specific tasks include providing guidance to INTEX by identifying discrepancies in satellite observations with (1) *in situ* measurements, (2) bottom-up emission inventories of nitrogen oxides and volatile organic compounds, and (3) model calculations of the export of pollution from North America to the global atmosphere. An important focus area is developing and improving bottom-up emission inventories by combining top-down and bottom-up information.

2. Year 1 Accomplishments

2.1. *Evaluation of GOME satellite measurements of tropospheric NO_2 and HCHO using regional data from aircraft campaigns in the southeastern United States*

In *Martin et al.*, [2004a] we compared tropospheric measurements of nitrogen dioxide (NO_2) and formaldehyde (HCHO) from the Global Ozone Monitoring Experiment (GOME) satellite instrument with *in situ* measurements over eastern Texas and the southeast United States. On average the GOME and *in situ* measurements of tropospheric NO_2 and HCHO columns are consistent despite pronounced sampling differences. The geometric mean *in situ* to GOME ratios over the campaign are 1.08 for NO_2 and 0.84 for HCHO, with corresponding geometric standard deviations of 1.27 and 1.38. The correlation of the observed column spatial variability between the two NO_2 measurement sets is encouraging before (r^2=0.54, n=18) and after (r^2=0.67, n=18) correcting for a sampling bias. Mean relative vertical profiles of HCHO and NO_2 calculated with a global 3-D model (GEOS-CHEM) and used in the GOME retrieval are highly consistent with *in situ* measurements; differences would affect the retrieved NO_2 and HCHO columns by a few percent. GOME HCHO columns over eastern Texas include contributions from anthropogenic volatile organic compound (VOC) emissions, but are dominated by biogenic VOC emissions at the regional scale in August-September when HCHO columns are within 20% of those over the southeastern United States. *In situ* measurements show that during summer the lowest 1500 m (the lower mixed layer) contains 75% of the tropospheric NO_2 column over Houston and Nashville, and 60% of the HCHO column over Houston. Future validation of space-based measurements of tropospheric NO_2 and HCHO

columns over polluted regions should include coincident *in situ* measurements that span the entire satellite footprint, especially in the heterogeneous mixed layer.

2.2 Variability of isoprene emissions over North America: comparing in situ observations and process models with top-down constraints from satellite observations of formaldehyde column

In *Palmer et al.*, [2004] we used formaldehyde (HCHO) column observations from the Global Ozone Monitoring Experiment (GOME) to test process-based models of VOC emissions from the terrestrial biosphere during the 2001 North American growing season. Emissions of isoprene, alpha- and beta-monoterpene, and methylbutenol (MBO) from the MEGAN and GEIA bottom-up emission models are used to drive the GEOS-CHEM global chemistry transport model. We use the Master Chemical Mechanism (MCM) to understand and quantify time-dependent HCHO yields from the oxidation of these biogenic trace gases, which are important for the interpretation of the GOME data. We use results from the MCM to parameterize the source of HCHO from the oxidation of monoterpenes and MBO, providing an upper limit to their contribution to HCHO columns over North America. We find that isoprene oxidation dominates the production of HCHO during summertime over North America, confirming previous work, and that the contribution from monoterpenes and MBO to HCHO columns is comparable to the fitting uncertainty of the GOME data, compromising the ability of GOME to provide constraints on emissions of these gases. We invert a modeled regression between isoprene emission and HCHO column to estimate isoprene emissions from GOME HCHO data. We evaluate the seasonal cycle of isoprene emissions estimated from GOME using in situ isoprene observations from the PROPHET and Harvard Forest sites. GOME captures many of the daily features observed by these *in situ* data, in addition to resolving the broad monthly-mean seasonal cycle of isoprene at these forest sites. We put the 2001 growing season into context by comparing it with other years in the GOME record (1996-2001). Observed HCHO columns exhibit a large interannual variability in the seasonal cycle, corroborated by EPA PAMS isoprene concentration data over Atlanta for the same period. Surface temperature explains more than 80% of this observed variation but its relationship to isoprene emission is notably different to that used in current emission models. There is also evidence of unknown additional factors that affect isoprene on a continental scale but are not included in current bottom-up VOC emission models.

2.3 Space-based diagnosis of surface ozone sensitivity to anthropogenic emissions

In *Martin et al.*, [2004b] we present a novel capability in satellite remote sensing with implications for air pollution control strategy. We show that the ratio of formaldehyde columns to tropospheric nitrogen dioxide columns is an indicator of the relative sensitivity of surface ozone to emissions of nitrogen oxides ($NO_x \equiv NO+NO_2$) and volatile organic compounds (VOCs). The diagnosis from these space-based observations is highly consistent with current understanding of surface ozone chemistry based on *in situ* observations. The satellite-derived ratios indicate that surface ozone is more sensitive to emissions of NO_x than of VOCs throughout most continental regions of the Northern Hemisphere during summer. Exceptions include Los Angeles and industrial areas of Germany. A seasonal transition occurs in the fall when surface ozone becomes less sensitive to NO_x and more sensitive to VOCs.

2.4 Satellite mapping of rain-induced nitric oxide emissions from soils

In *Jaeglé et al.,* [2004], we used space-based observations of NO_2 columns from the Global Ozone Monitoring Experiment (GOME) to map the spatial and seasonal variations of NO_x emissions over Africa during 2000. The GOME observations show not only enhanced tropospheric NO_2 columns from biomass burning during the dry season but also comparable enhancements from soil emissions during the rainy season over the Sahel. These soil emissions occur in strong pulses lasting 1–3 weeks following the onset of rain, and affect 3 million km2 of semiarid sub-Saharan savanna. Surface observations of NO_2 from the International Global Atmospheric Chemistry (IGAC)/Deposition of Biochemically Important Trace Species (DEBITS)/Africa (IDAF) network over West Africa provide further evidence for a strong role for microbial soil sources. By combining inverse modeling of GOME NO_2 columns with space-based observations of fires, we estimate that soils contribute 3.3 ± 1.8 TgN/year, similar to the biomass burning source (3.8 ± 2.1 TgN/year), and thus account for 40% of surface NOx emissions over Africa. Extrapolating to all the tropics, we estimate a 7.3 TgN/year biogenic soil source, which is a factor of 2 larger compared to model-based inventories but agrees with observation based inventories. These large soil NOx emissions are likely to significantly contribute to the ozone enhancement originating from tropical Africa.

2.5 Retrieval of tropospheric NO_2 columns from SCIAMACHY in support of INTEX

We have retrieved tropospheric NO_2 columns from SCIAMACHY for the INTEX time period. The attributes of our retrieval include a sophisticated algorithm for spectral fitting, and particular attention to the air mass factor calculation using spatially varying surface reflectivity, a cloud correction based on FRESCO cloud fields, a correction for the temperature sensitivity of the NO_2 cross section using GEOS meteorological fields, and GEOS-CHEM shape factors. We are providing these NO_2 fields to the INTEX database.

2.6 Validation of SCIAMACHY satellite measurements of tropospheric NO_2 columns with aircraft measurements as part of INTEX

We are comparing space-based measurements of tropospheric NO_2 with TD-LIF (Thermal Dissociation - Laser Induced Fluorescence) aircraft measurements taken during the INTEX-North America campaign in July and August of 2004. We are integrating the vertical profiles measured by TD-LIF that are coincident in space and time with a SCIAMACHY ground pixel. We have used TD-LIF 1 minute averages for this purpose. Two different methods are used to find the residual VCD in the troposphere from SCIAMACHY data: limb-nadir matching and the reference sector method.

2.7 Developing a 2004 bottom-up inventory for North America

We have been developing a 2004 updated inventory for use in the model comparison and data assimilation aspects of the project. The target period is March-September, 2004. For data sources, we are using updated approaches for various sources, and building upon an updated 2002 inventory by Hoyoux (recently developed and released, correcting errors in and updating the 1999 NEI), which we believe is the best foundation for the period and domain of interest. At

present, we are both processing emissions for the target period and running MM5 (which is needed for the detailed inventories, as some of the sources depend upon meteorological variables). Advances to the data assimilation process are also on-going.

3. Plans for Year 2

Our plans for Year 2 include the following:

1. Complete the validation of SCIAMACHY satellite measurements of NO_2 with INTEX aircraft measurements
2. Combine top-down NO_x emission information from SCIAMACHY with the bottom-up NO_x emission inventory over North America
3. Begin regional modeling of summer 2004
4. Examine the outflow of NO_x into the North Atlantic ocean

4. Publications Acknowledging Support From NNG04GE15G

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