# Chemical Modeling for studies of GeoTRACE capabilities

*Abstract.*

Geostationary measurements of tropospheric pollutants with high spatial and temporal resolution will revolutionize the understanding and predictions of the chemically linked global pollutants aerosols and ozone. However, the capabilities of proposed geostationary instruments, particularly GeoTRACE, have not been thoroughly studied with model simulations. Such model simulations are important to answer the questions and allay the concerns that have been expressed in the atmospheric sciences community about the feasibility of such measurements. We proposed a suite of chemical transport model simulations using the EPA Models 3 chemical transport model, which obtains its meteorology from the MM-5 mesoscale model. The model output consists of gridded abundances of chemical pollutants and meteorological parameters every 30-60 minutes for cases that have occurred in the Eastern United States. This output was intended to be used to test the GeoTRACE capability to retrieve the tropospheric columns of these pollutants.

## Project description

Understanding the roles of tropospheric ozone ($O_3$) and aerosols in perturbing the Earth system is a research priority for NASA and the U.S. Global Change Research Program (USGCRP). A major challenge for the USGCRP is determining the controlling chemical and transport processes for tropospheric $O_3$ and aerosols. Geostationary measurements of tropospheric ozone and its precursors with high spatial and temporal resolution can address four critical questions.

- Q1: What are the spatial and temporal emission patterns of the precursor chemicals for ozone and aerosols?
- Q2: What are the influences of weather in transforming and dispersing emissions, ozone, and aerosols into the global pollution?
- Q3: What is the evolution of the pollutants ozone and aerosols, from chemical formation and transport to losses by chemistry and deposition?
- Q4: What are the regional budgets for carbon monoxide, ozone, and aerosols over North America and regions bordering the Indian Ocean?

GeoTRACE enables answers to these science questions by measuring, from geostationary orbit, the tropospheric column densities of chemically linked nitrogen dioxide ($NO_2$), formaldehyde ($CH_2O$), carbon monoxide (CO), ozone ($O_3$), and scattering and absorbing aerosols (i.e., aerosol optical depth and single scattering albedo). Measured cloud heights help distinguish constituents in the planetary boundary layer (PBL) (< 3 km) from those in the free troposphere (FT). These constituents are measured every half hour during daylight with about 5 km horizontal resolution. The field-of-view is 5000 km × 5000 km.

GeoTRACE was proposed for Earth System Science Pathfinder Announcement of Opportunity AO-01-OES-01 last year. Unfortunately, it was not selected to go on to Step

2 of the proposal process. While the review panel agreed with the importance of the measurements, the proposal just didn't fire the imaginations of either the review panel or the program administrators. A more detailed discussion with program administrators revealed a number of objections

*Answers to the objections raised by the review process.*

- **High vertical resolution is required for studying tropospheric chemistry.** The important plumes are the PBL plumes downwind of urban, industrial, and power-generating areas and the large, thick free tropospheric plumes from biomass-burning, forest fire, and lofted industrial plumes. Species are generally well-mixed within these plumes. For quantifying emissions, the total tropospheric column is what is important.

- **Process studies cannot be done from space.** Transport processes are clearly possible by combining GeoTRACE information with weather information. Measuring the decay of primary emissions and emergence of resulting pollutants allows measurement of the time constants for $O_3$ and aerosol production from emissions, i.e., the chemistry. Linking these thousands of observations to the intensive field studies that put aircraft into plumes provides an unprecedented view of tropospheric pollution processes.

- **Satellite measurements of tropospheric chemistry are useless in regions where no good meteorology or ground-based pollution networks exist.** Once satellite measurements are thoroughly tested in a region with networks for meteorology and emissions, those satellite measurements will *enable* the understanding of pollution and its effects in essentially unsampled regions of the world, such as Africa and the Indian subcontinent, which may be contributing significantly to the global pollution.

- **Ground-based networks already give us all the information that we need about emissions.** Intensive field studies show huge errors (factors of 2 to 7, or more) in reported emissions inventories of photochemically important trace species. Models using emission inventories fail to reproduce the observed large values of $O_3$. Ground-based networks are not dense enough to capture all emissions and pollutant production for urban areas, much less regions and continents.

- **Only $CO_2$ matters for global climate change.** James E. Hansen and Makiko Sato (Trends of measured climate forcing agents, *Proc. Natl. Acad. Sci. USA*, Vol. 98, 14778-14783, 2001) state: "Climate forcing by $O_3$ and BC aerosols combined may be comparable to that of $CO_2$, yet neither constituent is included in the Kyoto Protocol. Some reduction in air pollution may occur as an incidental co-benefit of reduced fossil fuel use, but the small Kyoto emission reduction may be unnoticeable. Much larger reductions of air pollution are feasible. Achievement requires a strategy that explicitly targets $O_3$ and BC." $CO_2$ reduction is still necessary, but reducing $O_3$ and BC aerosols has simultaneous health benefits.

These objections are best answered by chemical transport modeling studies. Time evolving gridded model output is needed for select pollutants in order to test the retrievals and potential impacts of geostationary satellite measurements of these pollutants.

### Research strategy

This product of this proposal is output from the model runs specified in Table 1. The gridded model output will then be used by GSFC scientists to model the signals that the GeoTRACE satellite instruments will see and then to retrieve the tropospheric gas abundances for $O_3$, CO, $NO_2$, HCHO, $SO_2$, and scattering and absorbing aerosol.

| Table 1. Proposal product characteristics | |
|---|---|
| pollutants (on grid) | $O_3$, CO, $NO_2$, HCHO, $SO_2$, aerosol (scattering and absorbing) |
| meteorology (on grid) | pressure, temperature, cloud fraction, solar irradiance |
| horizontal resolution | 3 resolutions: 4 km x 4 km; 36 km x 36 km; 100 km x 100 km |
| vertical resolution | troposphere: 2 km; stratosphere: 5 km |
| horizontal coverage | minimum: 500 km x 500 km |
| vertical coverage | surface through stratosphere |
| temporal resolution | 30 minutes during the day |
| temporal duration | 2-3 cases; minimum: 36 hours; maximum: 5 days |
| model run variants | TBD, but the following scenarios are possibilities:<br>emission inventory, ½ x & 2 x isoprene;<br>emission inventory, ½ x & 2 x $NO_2$;<br>emission inventory, ½ x & 2 x urban carbon<br>emission inventory, 1/10 x & 10 x aerosol |
| main cases | summertime East Coast with high pressure (pollutants in PBL)<br>spring/fall East Coast frontal passage (pollutants lofted, clouds) |
| format | TBD, but netCDF a possibility |

The chemical transport model will be run by Dr. Ariel Stein, who works at NOAA, currently as an employee of the Pennsylvania State University. He will use the EPA Models 3 chemical transport model for the chemical runs. The meteorology will come from output of the Penn State/NCAR MM-5 mesoscale model (Figure 1).
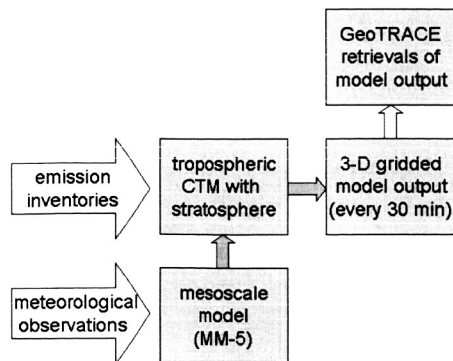
Figure 1. Flow chart of modeling project. This proposal's product will be the 3-D gridded model output of pollutant mixing ratios for every 30 minutes during 2 or 3 events, lasting from 1 to 4 days.

### *Research Activity and Results.*

When funding began, Ariel Stein purchased the proposed computer system and installed CMAQ and its components on it. Some parts were difficult to install and get working, but within a few months, CMAQ was operational.

In a meeting with Nelson Seaman and David Stauffer in September 2003, three cases were discussed. These cases were essentially already available in MM5 for 32, 12 and 4 km resolution are NARSTO case on12-15 July 1995, five days in July 1990, and CAPTEX in September 1983. The first two cases are stagnant air cases with high pollution. Augmented measurements were made during those periods. The CAPTEX case was a rapid synoptic case with warm conveyor belt lifting. Two cases of the three were chosen: the NARSTO case in 1995 and the CAPTEX case in 1983. The sensitivity scenarios were also chosen. In April 2003, computer model runs related to the NARSTO summer case from 1995 were submitted to NASA GSFC for use for simulating the satellite measurements. The runs comprise the following sensitivity analyses:

| Table 1. Competed CMAQ model runs | | | |
|---|---|---|---|
| case/resolution | 36 km | 12 km | 4 km |
| base | done | done | done |
| $0.5 \times NO_x$ | done | done | done |
| $2 \times NO_x$ | done | done | done |
| $0.5 \times VOC$ | done | done | done |
| $2 \times VOC$ | done | done | done |
| $0.5 \times Isoprene$ | done | done | done |
| $2 \times Isoprene$ | done | done | done |
| $0.5 \times SO_2$ | done | done | done |
| $2 \times SO_2$ | done | done | done |

Unfortunately, shortly after these runs were completed, Ariel Stein took a job in Spain. A Penn State graduate student was enlisted to carry on the work but was unable to complete it due to health problems. The completed model runs reside at GSFC. Fortunately, another graduate student is now making progress on the modeling and should be able to devote some of his time to completing this project.

An example of what can be learned can be seen in a demonstration case for CMAQ, which is centered on middle Tennessee. Four graphs are shown for each of ozone, $NO_2$, CO, and HCHO. In each set of four graphs, upper left is the surface values, upper right is the column in the planetary boundary layer (assuming 0-1.5 km), lower left is the free tropospheric (FT) column (assuming 1.5-8 km), and lower right is the total tropospheric column. The values for 10 am are shown.
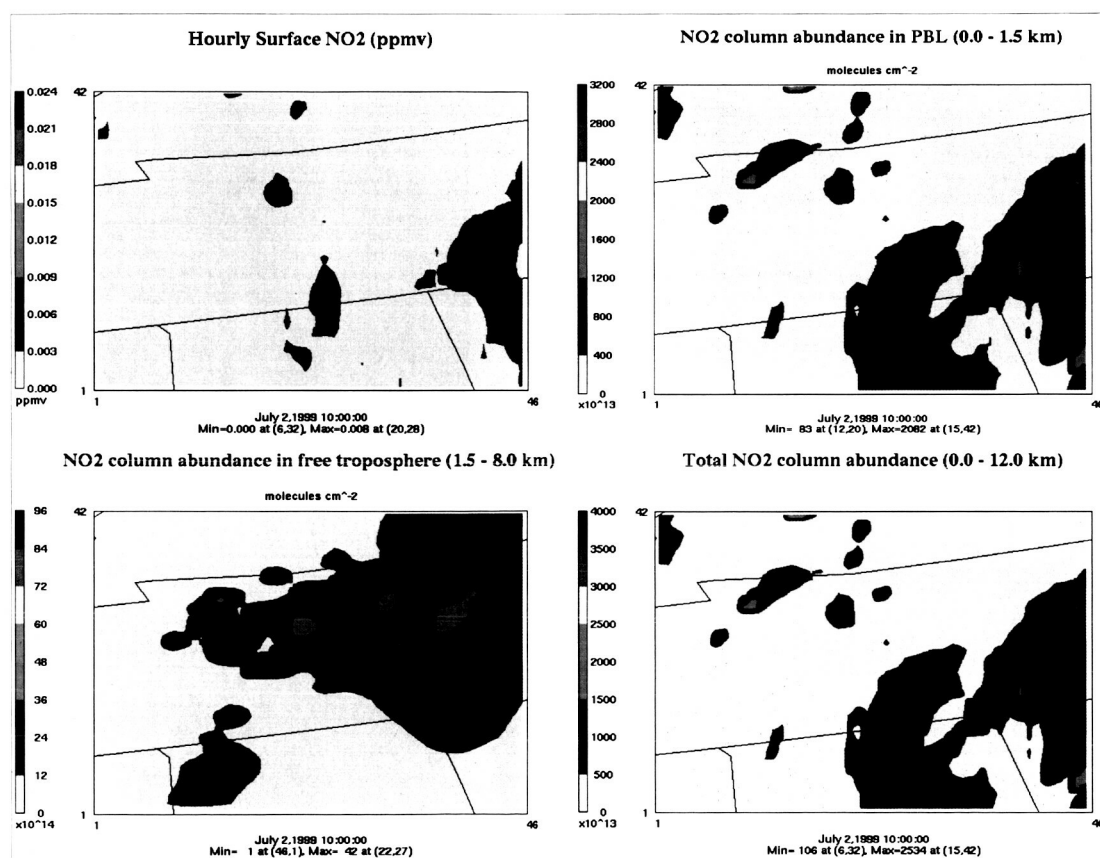


Figure 2. Nitrogen dioxide from CMAQ. Surface values are 0 to 24 ppbv, column values are for the PBL 0-320 $\times 10^{14}$ molecules cm$^{-2}$, for the Free Troposphere 0-96 $\times 10^{14}$ molecules cm$^{-2}$, and for the total tropospheric column 0-400 $\times 10^{14}$ molecules cm$^{-2}$.

For nitrogen dioxide, the PBL column is much greater than the FT column (Figure 2). This dominance of the PBL column over the FT column is illustrated by the close resemblance of the PBL column and the total tropospheric column. The $NO_2$ emissions

from Nashville (north central Tennessee) and Chattanooga (southeast Tennessee near the Georgia border) are evident in the both the surface and the PBL column maps. Several of the other enhanced $NO_2$ that appear in the PBL plot but not the surface plot are emissions from fossil fuel fired power plants. The large Cumberland power plant is located just to the west of Nashville.

For carbon monoxide, the PBL column is also much greater than the FT column for this case (Figure 3). Nashville and Chattanooga are marked by enhanced CO in both the surface and the PBL plots. Note the absence of emissions from the power plants. The longer lifetime for CO compared to $NO_2$ results in a more uniform distribution for CO than $NO_2$, with intense sources embedded in the broad distribution.
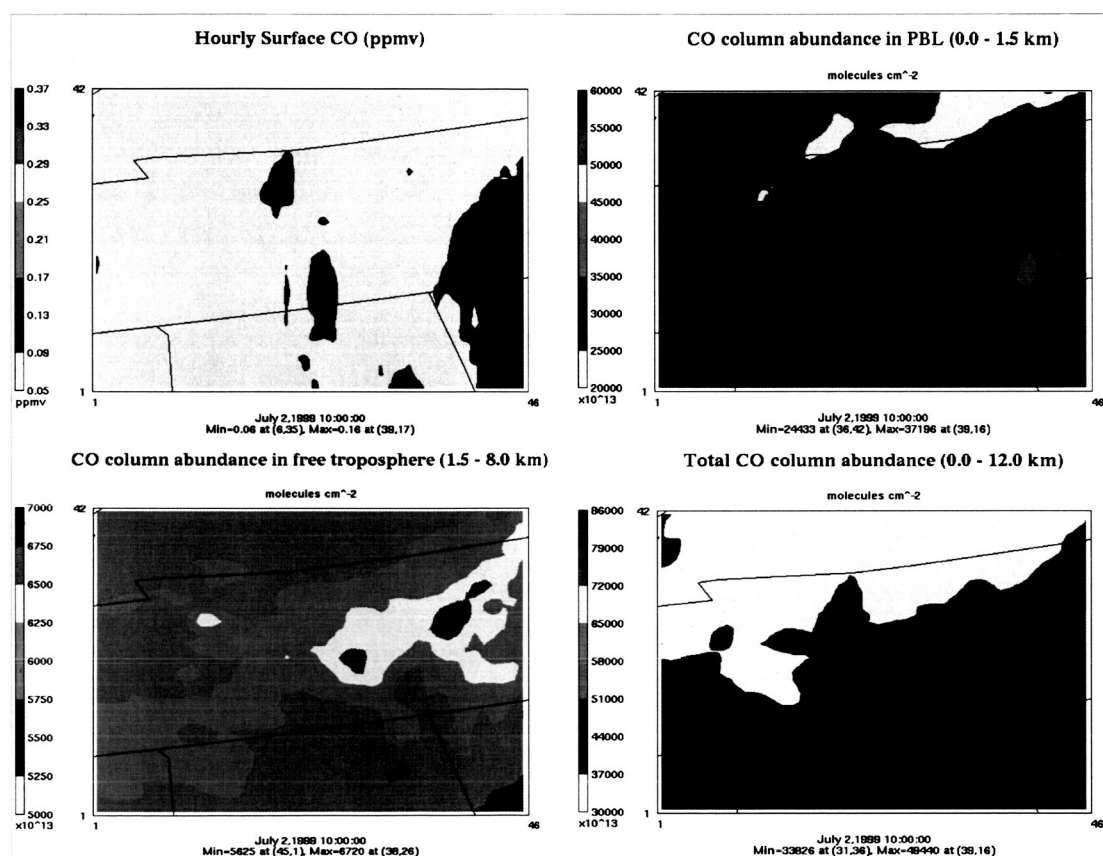


Figure 3. Carbon monoxide from CMAQ. Surface values are 0.005 to 0.370 ppmv, column values are for the PBL 2.0-6.0 x$10^{17}$ molecules cm$^{-2}$, for the Free Troposphere 0.50-0.70 x $10^{17}$ molecules cm$^{-2}$, and for the total tropospheric column 3.0-8.6 x $10^{17}$ molecules cm$^{-2}$.

For formaldehyde, the PBL column is ten times the free tropospheric column (Figure 4). HCHO has both biogenic and anthropogenic sources, although the HCHO for Nashville does not appear until the afternoon. The broad swath of HCHO is primarily from forests.

Later in the day, as the temperatures rise, so do the isoprene emissions from trees. Since isoprene reacts rapidly with OH, which is more plentiful at midday, the HCHO that is an oxidation product of isoprene increases rapidly.
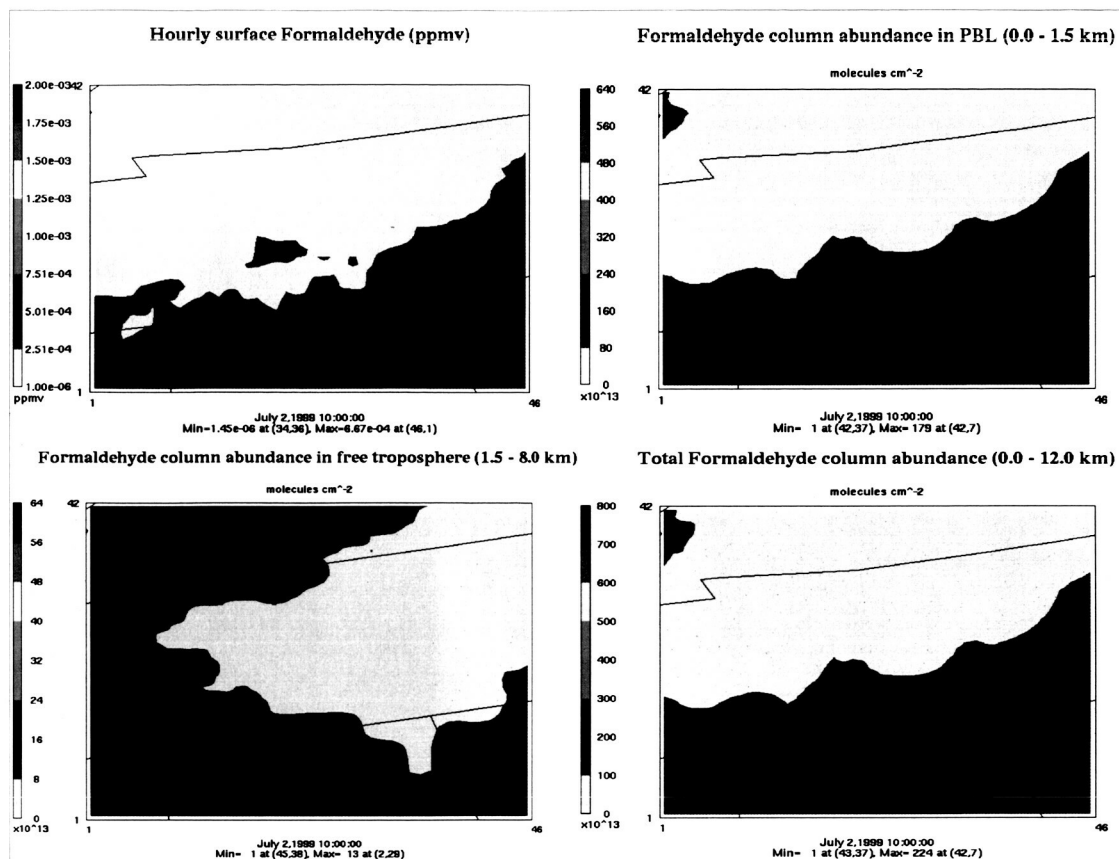


Figure 4. Formaldehyde from CMAQ. Surface values are 0.001 to 2 ppbv, column values are for the PBL 0 - 64 x$10^{14}$ molecules cm$^{-2}$, for the Free Troposphere 0 – 6.4 x $10^{14}$ molecules cm$^{-2}$, and for the total tropospheric column 0 - 80 x $10^{14}$ molecules cm$^{-2}$.

For ozone, the free troposphere contributes a few times more ozone to the column than the PBL does for this case (Figure 5). As a result, the total column's ozone pattern does not mirror either the free tropospheric or the PBL column faithfully. For this time on this day, neither Nashville nor Chattanooga has enhanced ozone that can be distinguished from the background regions.

While these plots from this one CMAQ case show the potential for geostationary measurements of tropospheric pollutants, they cannot capture the full power of them. The movies from which these plots were taken contain much more information.

All the proposed simulations for the most important case have been completed. A graduate student now has the capability to produce additional cases and to derive column information from CMAQ runs. The goal now is to produce the columns, observe them with a simulated geostationary instrument, and then retrieve the information.
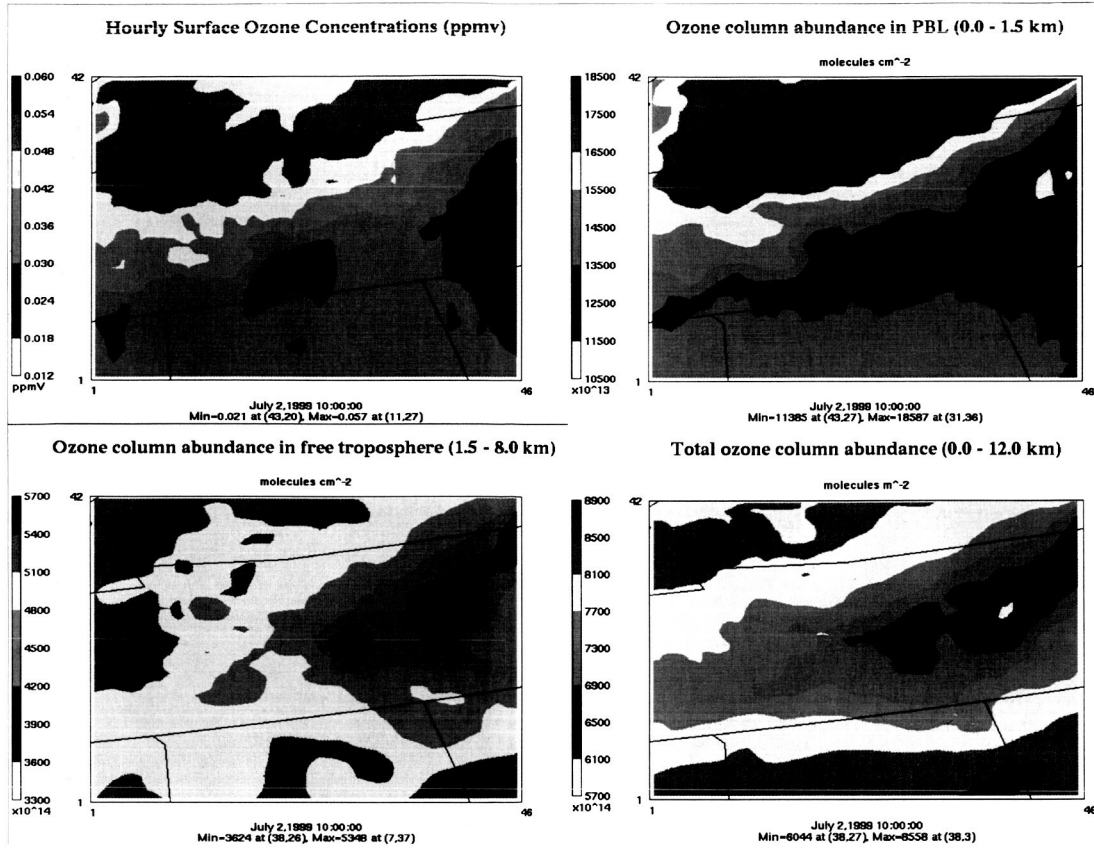


Figure 4. Ozone from CMAQ. Surface values are 0 to 0.060 ppmv, column values are for the PBL 0-1.850 x$10^{17}$ molecules cm$^{-2}$, for the Free Troposphere 3.30-5.70 x $10^{17}$ molecules cm$^{-2}$, and for the total tropospheric column 5.70-8.90 x $10^{17}$ molecules cm$^{-2}$.

## Publications.

none yet.