Advancing Emissions Quantification Techniques through the NASA AQAST Program

David G. Streets
Argonne National Laboratory, DIS/221
9700 South Cass Avenue, Argonne, IL 60439
dstreets@anl.gov

ABSTRACT

In many parts of the world and for many chemical species, emission inventories are not as reliable as we would like. Now that satellites can detect pollution over wide areas with daily resolution, the question arises as to whether the connection between facility or area emissions and satellite retrievals is robust enough to enable the retrievals to be used to infer emissions. This is one topic to be addressed by NASA's new Air Quality Applied Sciences Team (AQAST), a group of researchers convened to address how Earth science research and tools can be applied to air quality management. Satellites now make routine observations of many species associated with atmospheric pollution (O_3, NO_2, SO_2, CO, PM, HCHO, etc.). Studies have associated these column retrievals with bottom-up emissions, both with and without intervening model calculations of column amounts. The Ozone Monitoring Instrument (OMI) on the Aura satellite provides one of the most widely used retrievals. Studies of strong emitting sources around the world have revealed great potential for aiding the characterization of NO_2 and SO_2 emissions. A number of other applications may prove valuable: problematic industrial complexes, uncertain area sources (including biogenic), verification of regional emission reductions, quantification of uncertain Mexican and Canadian (and global) emissions and their importance for pollution import to the U.S., etc. Initially, AQAST will draw together researchers and EPA staff to review existing studies and capabilities and assess their potential use in air quality management, ultimately focusing on development needs for the most promising research directions.

INTRODUCTION

Last year NASA convened a new Air Quality Applied Sciences Team (AQAST) to focus on specific applied research topics required to advance the air quality management community's use and application of NASA Earth science observations and models in decision making. It was intended that the capabilities of the team would span all relevant NASA satellite mission observations and include data products from non-NASA satellites, including foreign satellites. The structure of the Air Quality Applied Sciences Team includes members broadly representative of the air quality community. Areas of expertise include:

- Regional atmospheric chemical and transport modeling capability as applied to air quality issues;
- Air quality forecasting;
- Emissions and emission inventory evaluation and improvement;
- Weight of evidence in air quality management decisions;
- Technical aspects of national and international air quality policy, including long-range transport, air quality management approaches in the U.S. and other countries, evolving policy needs arising from interactions between air pollution and climate change, and the national and international organizations that address air quality;
- Assessment of the information content of satellite observations by comparisons with data from other satellite sensors, calibration/validation activities, and well-calibrated existing ground-based sensors;

- Evaluation and use of air quality ground-based measurement networks;
- Analysis and modeling of satellite data to better characterize air quality observables and improve the representation of air quality observables in chemical transport models and climate models;
- Data services specifically for air quality data users, including archiving, sub-setting, remapping, and delivery;
- Air quality trends, compliance, policy analysis, policy efficacy, and accountability; and
- Economic valuations and socioeconomic benefits analysis techniques.

It was intended that the team would conduct new, focused, applied science activities on emerging and urgent needs in the air quality community. Typical activities include: developing new or significantly improving existing Level 2 or higher data products from the NASA instruments; providing technical feedback on NASA data products from the applications-oriented perspective and end users in the air quality community; developing applied research studies that scope and enable applications of Earth science products in air-quality decision making; using multiple satellite observations (especially multi-sensor products) to better connect research knowledge and results with air quality monitoring and decision-making activities of other Government agencies, businesses, and other organizations; articulating specific applied research needs and actual or possible societal benefits stemming from the research and applications; and integrating expertise on natural sciences, engineering, data systems, social sciences, and human factors to enable applications to organizational decision making. In this paper, we present new information on the application of satellite data and techniques to begin to help in the quantification of emissions from individual point sources.

BODY

Satellites can now make routine observations of many species associated with atmospheric pollution (O_3, NO_2, SO_2, CO, PM, HCHO, etc.). Studies have associated these column retrievals with bottom-up emission estimates, both with and without intervening model calculations of column amounts. The Ozone Monitoring Instrument (OMI) on the Aura satellite provides one of the most widely used retrievals these days. Studies of strong emitting sources around the world have revealed great potential for aiding the characterization of various source types, particularly for NO_2 and SO_2. The jury is still out, so to speak, on how useful this capability is for U.S. air quality management. For instance, the use of continuous emission monitors (CEMs) on U.S. power plants obviates, for the most part, the need to double-check emissions from these sources. Nevertheless, a number of other applications may prove valuable: problematic industrial sources and complexes, uncertain area sources (including biogenic), verification of regional emission reductions, quantification of atmospheric lifetimes, quantification of uncertain Mexican and Canadian emissions and their importance for pollution import to the U.S., coordinated use of multi-species retrievals, etc.

If the objective is to estimate the emissions from an anthropogenic point source—as distinct from a natural point source such as a volcano or an anthropogenic area source like the ammonia emissions from an agricultural region—then one would ideally like to study new individual sources of emissions where the source strength is sufficiently large. While work has been undertaken on the SO_2 emissions from metal smelters around the world[1,2] there are insufficient of them to undertake a comprehensive analysis. Coal-fired power plants would be a better option. At present, China and India represent ideal opportunities to demonstrate quantitative linkages between satellite retrievals and emission estimates. This is because of the ongoing construction of many large coal-fired power plants in relatively remote parts of these countries, away from other sources of emissions (transportation, industry, residential, etc.). Thus, we see large sources of emissions suddenly appear where there were previously only very low levels of emissions. These large emissions can be quantified from knowledge of activity levels in bottom-up emission inventories, and the emissions are readily observable by the new generation of satellite instruments. Such an analysis is not as easy in a place like the U.S., where we commission no large new sources of emissions these days, and existing emissions tend to be low and for the most part unchanging. As a place to begin, therefore, we can try to develop relationships in countries like China and India and then transfer the applications to the U.S.

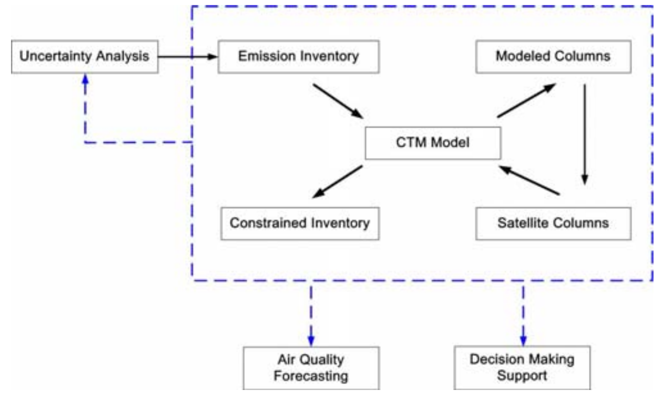

Figure 1 Schematic diagram of the combined use of emission inventories, observed satellite columns, and chemical transport models to improve emission inventories and benefit air quality analysis.

Comparing emissions directly with satellite columns is an incomplete connection, at best. The column observed by the satellite overpass is a composite of (a) newly emitted species at ground level, (b) species in transit from upwind sources, and (c) conversion products from upwind chemical reactions. The latter two may be present at high altitudes, which is important because the satellite is looking through an entire vertical column of air. The best way to make these inter-comparisons is to add a third element: modeled columns. If the bottom-up emissions are put into a regional chemical transport model that incorporates prevailing meteorology and chemical reactions, we should be able to calculate modeled columns that would agree better with satellite columns. This should all be performed within an uncertainty analysis framework to explicitly represent the confidence we have in each of the controlling components (see Figure 1).

In 2005 a groundbreaking paper by Richter et al. in *Nature*[3] reported the first assessment of emission changes in China from tropospheric NO_2 columns retrieved from GOME and SCIAMACHY satellite observations. This work was extended[4] to reconcile trends from satellite observations with the trends from bottom-up inventories for the period 1996-2004 (Figure 2). The fascinating analysis concluded that good agreement was achieved for summertime, but that the bottom-up trend was lower than the satellite trend in wintertime (Figure 3). Both techniques are known to present difficulties in winter—the bottom-up method due to uncertainty about the extent of home heating in colder latitudes and the satellite method because of possible contamination of the NO_2 signal with sulfate aerosol, which is high in wintertime. It was nevertheless recognized that China is a fertile place for studying the relationship between satellite retrievals and emission inventories, due to the large increases in emissions that occur when new power plants are commissioned there. The focus of attention switched to the Chinese region of Inner Mongolia—where phenomenal growth in coal-fired power generation has occurred since 2005. A comparison of three OMI retrieval products for monthly columns in 0.125º × 0.125º grids around new power plants with expected NO_x emission increases from inventories yielded very good agreement[5], potentially signaling good prospects for utilizing satellite products in emission characterizations (Figure 4). This was confirmed when SO_2 emissions from the same set of power plants was studied, rather than NO_x. Though the SO_2 columns are two to three orders of magnitude weaker than the NO_2 columns, the signal from a strongly emitting power plant is

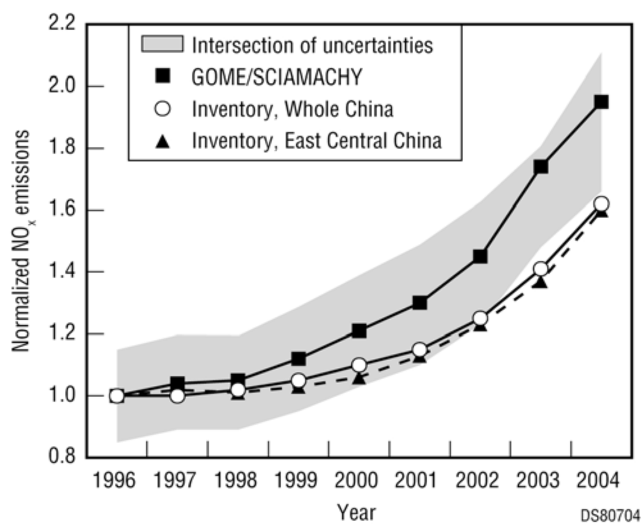

Figure 2 Trends of NO_x emissions in China from bottom-up inventories are lower than GOME or SCIAMACHY satellite NO_2 retrievals, but both sets of trends lie within joint uncertainty bounds.

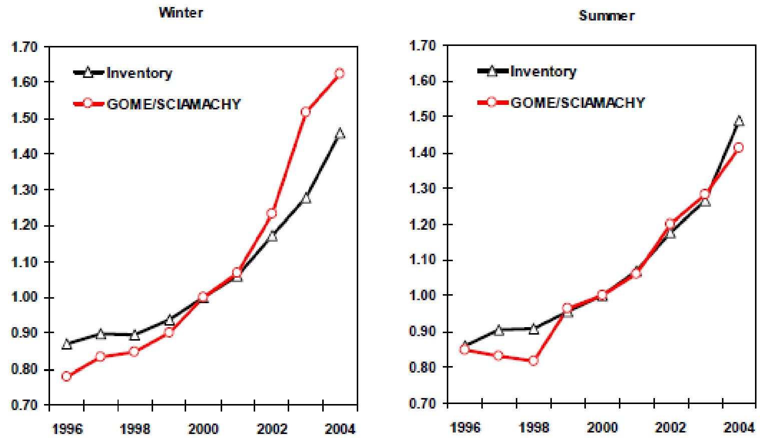

Figure 3 Comparison between GOME and SCIAMACHY NO_2 satellite retrieval trends over China with bottom-up inventories of NO_x in wintertime (satellite increase is greater) and in summertime (good agreement).

detectable. It was found possible to study SO_2 emissions at these plants and provide evidence as to whether they are complying with emission reduction requirements under a new Chinese law[6]. The potential power of such capability in the regulatory arena does not need to be spelled out. China's Ministry of Environmental Protection (MEP) has expressed great interest in this aspect of the work, and more details are presented below.

As was shown in Figure 1, to improve the association between bottom-up emission estimates and observed total column amounts, it can often prove beneficial to use a chemical transport model to simulate the column densities using the bottom-up inventory data and then compare the modeled columns with the satellite observations. To some extent, therefore, pollutant transport and chemical conversion are both taken into account. The next set of results examines the same region but makes the connection between bottom-up NO_x emission inventories for China and satellite NO_2 columns, using the GEOS-Chem regional model as an intermediary[7]. Figure 5 (left frame) shows NO_2 column distributions from the NASA OMI product for eastern China for JJA of 2007. A new national NO_x emission inventory for eastern China was assembled for this work, using confidential coal-fired power plant inventories provided by China's MEP, supplemented with other Argonne inventories for non-power sectors. This inventory was input into the Harvard GEOS-Chem model to predict NO_2 columns over the same region for the same time period (Figure 5, right frame). The resolution of the satellite column product is a little better (0.25º × 0.25º) than the model product (0.5º × 0.67º). Though the spatial patterns are similar (high values around Beijing/Tianjin, Shanghai, and Guangzhou), NO_2 columns are underestimated in the model at the present time. This may be an emissions issue, a model boundary/IC issue, a retrieval issue, or something else. More work is needed to improve these inter-comparisons. Unfortunately, we do not have an extensive ground-station network of NO_x concentration monitors in China (outside of the major cities, at least) for calibration. However, collaboration with China's MEP to obtain raw data from their monitoring network should provide an additional constraint on the GEOS-Chem model performance.

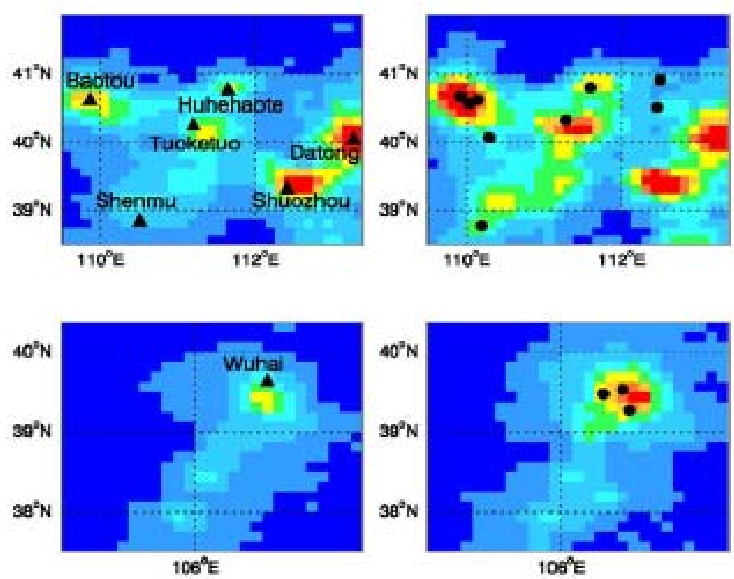

Figure 4 OMI average NO_2 columns over two regions of Inner Mongolia, China, for 2005 (left) and 2007 (right), where solid circles show the locations of new power plants built between 2005 and 2007 [from E-2]. Co-location is evident.

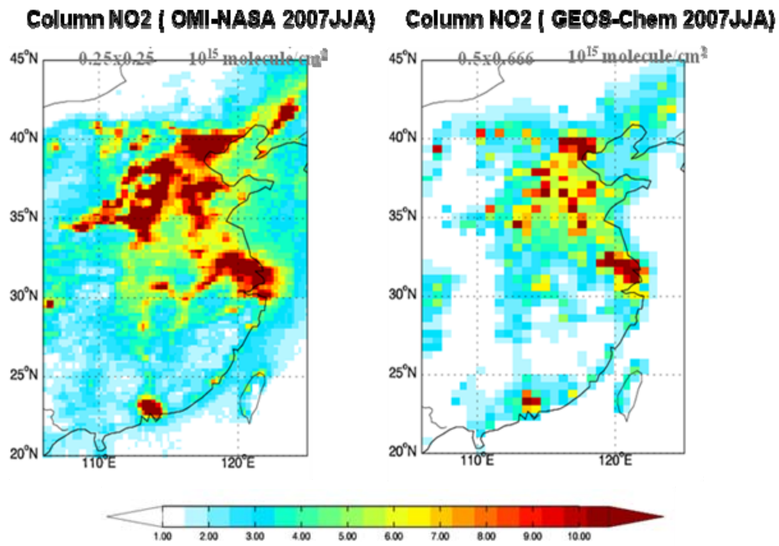

Figure 5 Comparison of OMI-NASA NO_2 columns with GEOS-Chem modeled NO_2 columns over eastern China for JJA, 2007.

When good region-wide agreement is reached between observed and modeled satellite columns, we will be in a position to focus in on any number of cities and power plants to develop robust quantitative relationships between the two. It is perhaps even more interesting to study SO_2 than NO_2. Control of SO_2 is a major priority for the Chinese government at present, and the installation of flue-gas desulfurization (FGD) equipment is now required (and enforced, supposedly) for all large new power plants. Retrofit of some existing plants is also underway. However, the SO_2 signal from OMI is two to three orders of magnitude weaker than the NO_2 signal and requires careful processing of the retrievals. Already, however, SO_2 columns have been processed by NASA for the same Inner Mongolian power plants previously studied. As for NO_2, we really need to look at China to find strong enough signals for SO_2; there are few large SO_2 sources any more in the U.S.

Figure 6 shows changes in summertime (JJA) average OMI SO_2 columns (line with open circles) for the period 2005-2008 for two specific power plants. The red bars are bottom-up inventory estimates assuming the plants are not using the required FGD devices, and the blue bars assume full usage of FGD devices. For the plant in Huhehaote, there is little evidence that the required FGD system began operating in 2007, as it was supposed to. On the other hand, we see confirmatory evidence of operation of the FGD system on the Shuozhou plant in 2008. Though this work is still in its exploratory stage, it is clear that we have sufficient spatial resolution and signal power in the OMI retrievals to monitor the emissions of major species from individual facilities.

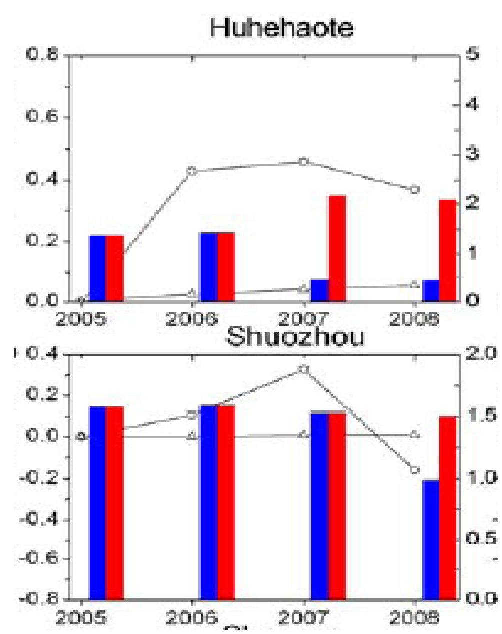

Figure 6 SO_2 emissions from two individual Chinese power plants over a four-year period.

It should also be possible to track temporal emission changes at the resolution of one day. Again, preliminary investigative research has been conducted to see if we can detect reduced NO_x emissions in China during the period of the Chinese Spring Festival celebrations (Figure 7). Normally, there is a large amount of traveling done prior to the festival, as people return to their family homes; but during the subsequent two weeks or so, NO_x emissions should be less than normal, as people are not traveling and factories are not operating. We have compiled OMI NO_2 retrievals for five years, 2005-2009. These must then be synchronized to the lunar calendar, because Spring Festival falls at different times of the year in the modern calendar. Using a five-day moving average to smooth out day-to-day variations that might correspond to interannual meteorological fluctuations, we find a distinct reduction of about 20% in the OMI NO_2 columns. The reductions begin during the three-day buildup to the Festival, continue for another 11 days or so, and then increase rapidly during the next two days, as people return to their current homes and begin work. We also see a smaller 2-3 day reduction after that, corresponding to the Lantern Festival (Yuan Xiao) that always occurs 15 days after the Spring Festival. This work is being expanded to other cities and time periods, to draw more general and firmer conclusions about observing anthropogenic activity changes from space.

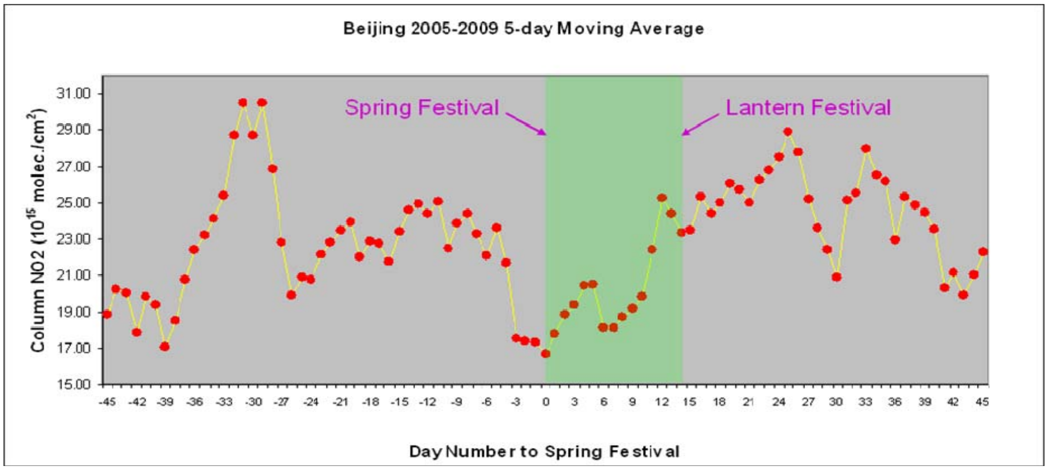

Figure 7 Five-day moving averages of daily OMI NO_2 retrievals over Beijing, averaged over a five-year period, synchronized to the Chinese celebrations of Spring Festival and Lantern Festival.

As a final step, a more comprehensive analytical approach was taken by incorporating the GEOS-Chem model into the analytical framework to simulate NO_2 columns over the whole of China[8]. The correlations of absolute changes between summertime average modeled NO_2 columns (ΔNO_2-*CTM*), OMI-retrieved NO_2 columns (ΔNO_2-*OMI*), and monthly average NO_x emissions (ΔNO_x) are shown in Figure 8 for selected regions of China.

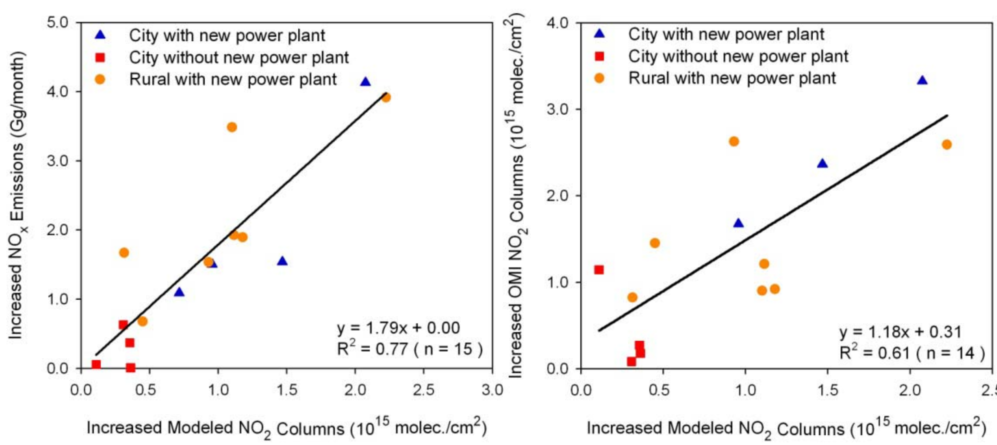

Figure 8 Scatter plots of increased modeled summertime average tropospheric NO_2 columns between 2005 and 2007 against increased summertime monthly average NO_x emissions (left) and increased OMI-retrieved summertime average tropospheric NO_2 columns (right) for selected sub-regions.

Good correlation was found between ΔNO_x and ΔNO_2-*CTM* ($R^2 = 0.77$), with a slope of 1.79, which indicates that increased NO_x emissions of 1.79 Gg/month will lead to increased local NO_2

columns of 1.0×10^{15} molecules/cm^2 in the summertime in these areas. ΔNO_2-*OMI* and ΔNO_2-*CTM* are also well correlated ($R^2 = 0.61$). The correlation between ΔNO_2-*CTM* and ΔNO_2-*OMI* ($R^2 = 0.61$, n = 14) in this work is of the same order of magnitude as the correlation between ΔNO_x and ΔNO_2-*OMI* ($R^2 = 0.68$, n = 9) in previous work[5]; however, comparisons in rural areas are much improved in this work, which means that we have a better representation of regional NO_x and the emissions from smaller sources through the use of the GEOS-Chem model. Because the estimated NO_x emissions in the studied areas are mainly contributed by newly built power plants, the emissions from which are well known, the consistency in ΔNO_2-*OMI*, ΔNO_2-*CTM,* and ΔNO_x means that the satellite measurements can be used to infer NO_x emissions from large point sources. It was recommended that further study should focus on the impact of newly added NO_x emissions from power plants on local NO_2 vertical profiles, which could play critical roles in the calculation of the air mass factors used to retrieve satellite measurements. We concluded from this entire body of work that dramatic increases of NO_2 columns during 2005-2007 in China, attributed to the construction of new power plants, can be readily observed by OMI. North China and Inner Mongolia showed the fastest increase rates of NO_2 columns in the country, followed by East China, Southwest China, and the southeast coastal regions.

Preliminary work has begun on examining whether a similar situation holds for India. There have been very few applications of satellite retrievals to India. Two previous papers[9,10] studied the interannual trends and seasonal variations of major NO_2 hotspots over India using NO_2 retrievals from GOME, SCIAMACHY, and OMI. However, they only gave a qualitative or semi-quantitative analysis of the relationship between NO_x emissions and satellite retrievals, partly due to the lack of reliable NO_x emission inventories for large point sources (LPS). We have examined the India NO_x emission trend of thermal power plants during the period 1996−2010 from the viewpoints of both unit-based inventories and multi-satellite observations[11].

Because of the large increase in the NO/NO_2 ratio with altitude and the short lifetime of NO_x in the lower mixed layer, land surface NO_x emissions and NO_2 columns are closely correlated, and to a first approximation, changes in NO_2 columns are expected to be proportional to changes in NO_x emissions. This makes it possible to identify the NO_x sources from NO_2 column maps. Figure 9 shows the spatial distribution of average NO_x emissions from thermal power plants and all-month average of NO_2 columns over India from four instruments. A number of satellite NO_2 hot spots (especially for OMI, SCIAMACHY, and GOME-2, which have finer footprints) are observed over India, and they match the locations and the amounts of NO_x emissions of thermal power plants well.

Influenced by the South Asia monsoon meteorology, India is largely subject to four seasons: winter (December−February), summer/pre-monsoon (March−June), monsoon (late June−September), and post-monsoon (October−November). NO_2 columns are high in winter and low in the monsoon season due to the seasonal variation of NO_x lifetime and the effective wet scavenging during the monsoon season. Three major factors impact the comparison between satellite NO_2 retrievals and emission estimates: uncertainty in the satellite retrievals, seasonal variation of NO_x lifetime, and incomplete conversion of NO to NO_2. These factors are the reasons why summer (June−August) NO_2 retrievals were always used in previous studies to quantitatively analyze NO_x emissions in the U.S. and China. The short NO_x lifetime, low snow cover, small solar zenith angle, and strong actinic flux in summer make the retrievals more reliable and the link between emissions and the NO_2 columns more direct than the other seasons in these two countries. However, for India, although the NO_x lifetime is short during the monsoon period, late June to September is the worst period to observe NO_2 from satellites. In India, more than 70% of the annual precipitation occurs in the monsoon season. The frequent cloud cover and heavy rainfall during this period results in insufficient satellite observation samples and extremely low NO_2 surface concentrations, which makes the satellite retrievals highly uncertain.

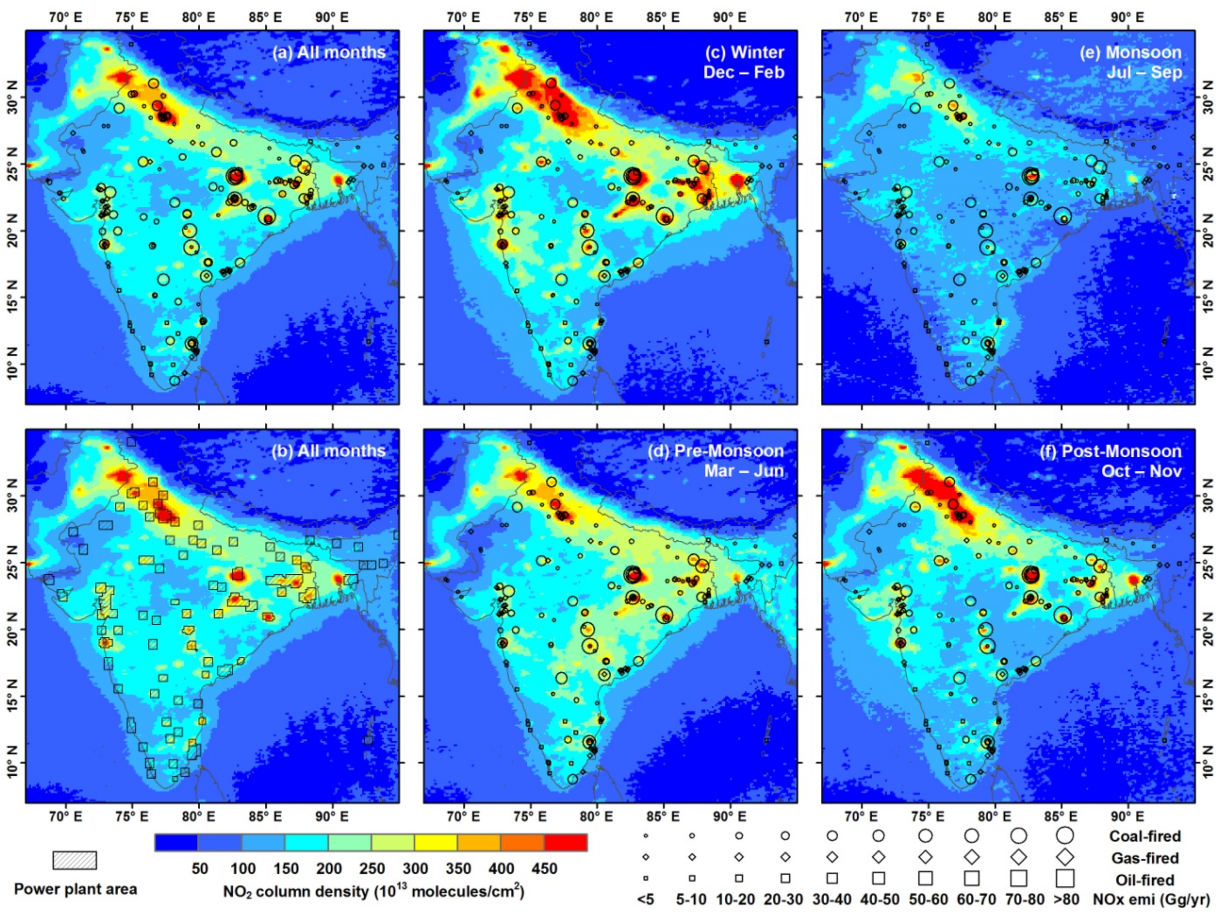

Figure 9 Spatial distribution of (a, c−f) average NOx emissions from thermal power plants, (b) defined power plant areas, and (a, b) annual and (c−f) seasonal average of OMI NO2 TVCDs over India during 2005−2010.

However, the fact that India is closer to the equator (the latitude is ~20º lower) than the U.S. and China brings tremendous advantages to India for the use of other months' NO_2 retrievals in the quantitative analysis. First, due to the low latitude, temperature and actinic flux are high in India, resulting in a relatively short lifetime of NO_x all year round. Second, low snow cover and favorable satellite measurement geometry at small solar zenith angle make the NO_2 retrievals very reliable. Third, compared to the U.S. and China, the seasonality of temperature, actinic flux, NO_x lifetime, and NO_2 retrievals in India is weaker, which means that all-year data can be used in the analysis and the NO_2 retrievals' uncertainties are further reduced. Taking into account all the above factors, we selected non-monsoon months (i.e., January−June and October−December) as our study period for India, in contrast to the summer NO_2 retrievals used in previous studies. An additional favorable condition for quantitative analysis in India is that area NO_x emissions are smaller and individual NO_x sources and NO_2 hotspots are more isolated and discernible all over the country (except for the region around Delhi) in contrast to the dense regional NO_2 plumes over the northeastern U.S. and the eastern central China. Work is ongoing to further probe the relationships between NO_2 satellite retrievals over India and bottom-up emission inventory estimates. The relationships developed for India will be compared with China and subsequently for Southeast Asia, which will be investigated as part of a separate NASA program, SEAC4RS. A synthesis of the results for the entire Asian region will then be reviewed for its relevance to understanding the emissions from sources in North America. Figure 10 shows combined OMI NO_2 retrievals averaged over the period 2005−2010 for a large swath of Asia, in relationship to the locations of individual power plants. Note that the intensity scales are different, emphasizing the much larger signal obtained for China than for India. Some of the summary data for the two countries are shown at right and left, indicating the promise of the technique.

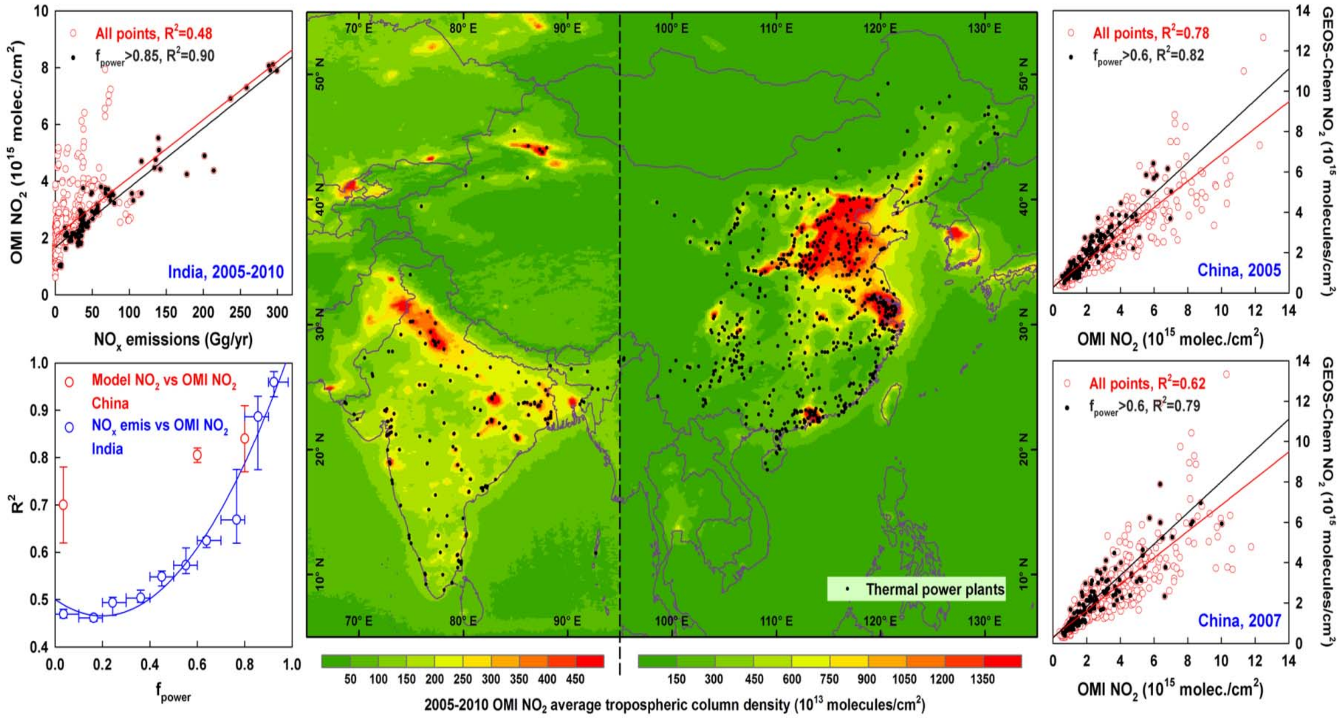

Figure 10 Average OMI NO_2 column densities over China and India for the period 2005-2010 in relationship to the locations of major thermal power plants, showing some statistical relationships between column densities and bottom-up emissions (left for India, right for China)

CONCLUSIONS

Though much information can be obtained by individual comparisons of emissions and retrievals, we believe that the ideal way forward to improve emission estimates and to use spatially and temporally refined information to aid compliance verification is to integrate findings from bottom-up emission inventories, satellite retrievals, field monitoring, and chemical transport modeling. Insights can be considerably enhanced (a) when multiple species can be examined synchronously; (b) when studies can be done across different human environments (e.g., urban vs. rural); and (c) when day-to-day and year-to-year variations can be studied. Such techniques as data assimilation and inverse modeling will also be beneficial. One day, we feel sure, the “eye in the sky” will be an important tool for compliance verification. NASA has taken an important step towards this goal by assembling the AQAST team and using team members to develop the connections between NASA scientists, who are assembling the tools and methods to use space-based observations, and U.S. air quality managers, who can use the resulting products to improve the efficiency and effectiveness of their analyses and, ultimately, their decisions.

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