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Evaluation of long-term tropospheric NO_2 data obtained by GOME over East Asia in 1996–2002

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[1] Long-term tropospheric nitrogen dioxide (NO_2) column data obtained by the Global Ozone Monitoring Experiment (GOME) (G-NO_2) are evaluated to confirm the trends found in tropospheric NO_2 abundances over East Asia between 1996 and 2002. For three locations in Central and East Asia, the G-NO_2 values are compared with tropospheric columns estimated from coincident observations of total NO_2 by ground-based UV/visible spectrometers and stratospheric NO_2 by satellite solar occultation sensors (E-NO_2). The comparisons show a slight linear drift in G-NO_2 data from 1996 to 2002. However, it is much smaller than the standard deviation of the differences between G-NO_2 and E-NO_2 and much smaller than the increasing trends in NO_2 seen by GOME over the industrial areas of China, demonstrating the validity of the trends estimated using the GOME data.
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1. Introduction

[2] It is well known that nitrogen oxides (NO_x = NO + NO_2) play a crucial role in the Earth's atmosphere. As a result of rapid economic growth in East Asian countries, an increase in the anthropogenic emissions of NO_x from these countries is thought to be occurring, due mainly to rising consumption of fossil fuels [*Akimoto*, 2003]. While tropospheric NO_x abundance should respond to the changes in emission from adjacent regions, its temporal evolution over East Asia has not yet been quantified due to the lack of continuous long-term observations by a single instrument. The Global Ozone Monitoring Experiment (GOME) onboard the European Remote Sensing-2 (ERS-2) satellite measured tropospheric NO_2 column abundances over East Asia from July 1995 until June 2003 [*Leue et al.*, 2001; *Richter and Burrows*, 2002; *Martin et al.*, 2002, 2003; *Beirle et al.*, 2004; A. Richter et al., Significant increase in nitrogen dioxide levels over China observed from space, submitted to *Nature*, 2005, hereinafter referred to as Richter et al., submitted manuscript, 2005]. Recently, Richter et al. (submitted manuscript, 2005) have combined the GOME tropospheric NO_2 column data (G-NO_2) with those of the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) to show a highly significant increase in NO_2 of ~50% over the industrial areas of China (30°–40°N and 110°–123°E) between 1996 and 2004. In Europe from 1996 to 2001, G-NO_2 has shown a reduction consistent with the pronounced reduction of NO_x emissions (Richter et al., submitted manuscript, 2005). G-NO_2 has been compared to independent measurements over Europe and the U.S. [e.g., *Heland et al.*, 2002; *Petritoli et al.*, 2004; *Martin et al.*, 2004], and good agreement was found. For East Asia, however, despite a large uncertainty in emission estimates, no validation comparison has been made so far. In the present study, the long-term G-NO_2 data obtained in Central and East Asia are evaluated by comparing monthly-mean G-NO_2 tropospheric column data with those estimated from long-term observations by ground-based UV/vis. zenith-sky spectrometers and satellite-borne solar occultation sensors (E-NO_2). The comparisons support the results of Richter et al. (submitted manuscript, 2005) indicating a significant increase in NO_2 over the industrial areas of China between 1996 and 2002.

2. GOME Tropospheric NO_2 Column (G-NO_2)

[3] The GOME instrument, a nadir-viewing UV/vis. spectrometer, was launched aboard the ERS-2 satellite in April 1995 [*Burrows et al.*, 1999]. GOME observations with a spatial resolution of 40 km (in latitude) × 320 km (in longitude) are made three times across the flight track using a scanning mirror, achieving nearly global coverage within 3 days. The satellite crosses the equator at 10:30 am local time in the descending node, so that GOME observations at northern mid-latitudes occur around 11:00 am. For the G-NO_2 tropospheric column data used in this study, the retrieval technique was described in detail by *Richter and Burrows* [2002] and recent improvements were given by Richter et al. (submitted manuscript, 2005). To minimize the shielding effects of clouds on the retrieval, we used GOME data with a cloud fraction less than 0.2, based on a cloud retrieval algorithm (FRESCO) [*Koelemeijer et al.*, 2001]. It has been shown that G-NO_2 tropospheric column data was tightly correlated with surface NO_2 concentration in highly-polluted regions (tropospheric NO_2 reaching 200 × 10^{14} cm^{-2}) in Italy throughout the year 2000 under optimized comparison conditions (well-mixed boundary layer

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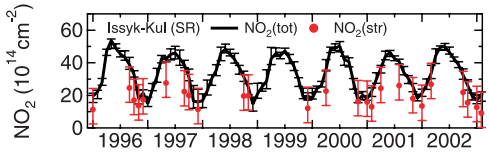

Figure 1. Time series of NO_2(tot) (black) and NO_2(str) (red) values for Issyk-Kul at sunrise (SR). The SAGE II- and HALOE-based NO_2(str) values have been unified for each month.

and low cloud coverage) [*Petritoli et al.*, 2004], indicating the sensitivity of the GOME observations to NO_2 near the surface. The present study uses monthly-averaged G-NO_2 data derived on a 0.5° (in latitude) × 0.5° (in longitude) grid. We focus on the years 1996–2002, when nearly complete seasonal cycles were observed by GOME. Uncertainty in G-NO_2 arises primarily due to errors in the a-priori NO_2 profiles used and the tropospheric air mass factor calculations. Uncertainty in a single G-NO_2 value was estimated to be $\sim 15 \times 10^{14}$ cm^{-2} [*Richter and Burrows*, 2002] or 35–60% under highly-polluted conditions [*Boersma et al.*, 2004], suggesting that there is much smaller uncertainty in the monthly-averaged G-NO_2 data.

3. Estimate of Tropospheric NO_2 Column (E-NO_2)

[4] To estimate E-NO_2 values for the period 1996–2002, we combine observations by ground-based UV/vis. zenith-sky spectrometers and satellite-borne solar occultation sensors (Stratospheric Aerosol and Gas Experiment II (SAGE II) and Halogen Occultation Experiment (HALOE)) with the Chemical Atmospheric general circulation model for Study of Atmospheric Environment and Radiative forcing (CHASER) [*Sudo et al.*, 2002]. E-NO_2 is defined as:

$$E\text{-}NO_2 = (NO_2(\text{tot}) - NO_2(\text{str})) \times F,$$

where NO_2(tot) is the total (troposphere + stratosphere) NO_2 vertical column derived from a UV/vis. spectrometer at a local time of sunrise (SR) or sunset (SS), NO_2(str) is the stratospheric NO_2 vertical column derived from SAGE II (ver. 6.2) and HALOE (ver. 19) observations at SR or SS, and F is a diurnal correction factor defined as the ratio of the modeled tropospheric NO_2 column at the time of the GOME measurements to that at SR or SS.

[5] For estimating E-NO_2 values, we focus on three sites, (1) Moshiri (44.4°N, 142.3°E, 200 m asl), Japan, (2) Issyk-Kul (42.6°N, 77.0°E, 1650 m asl), Kyrgyzstan, and (3) Zhigansk (66.8°N, 123.4°E, 50 m asl), Russia, where UV/vis. spectrometer observations at SR and SS were made as part of the Network for the Detection of Stratospheric Change (NDSC) during the 1996–2002 period. These sites are located in or closer to East Asia than other UV/vis. spectrometer observation sites available in the NDSC database for the period. We used the differential optical absorption spectroscopy (DOAS) technique and air mass factor for ground-based observations (AMF) to derive NO_2(tot) at SR and SS from the spectra measured. Constant AMF values of 17.0, 16.5, and 15.86 over 1996–2002 were used for Moshiri, Issyk-Kul, and Zhigansk, respectively. These AMF values were taken from the NDSC database. It should be noted that the use of constant AMF values may contribute to the uncertainty in NO_2(tot), because AMF can vary depending on season. It should also be noted that an additional uncertainty in NO_2(tot) may arise depending on season, because the sensitivity of the observations at SR and SS to the troposphere is much less than that to the stratosphere and varies according to the vertical profile of NO_2. Moreover, the retrievals of NO_2(tot) for the three sites have not been performed in a fully consistent manner, including the same NO_2 cross sections and a radiative transfer model. The effect of these uncertainties on the G-NO_2/E-NO_2 comparison is discussed in the results and discussion section below.

[6] We next selected stratospheric NO_2 vertical profiles observed by SAGE II/HALOE at SR or SS within 2° latitude, 500 km distance, and 1 day of the ground-based UV/vis. spectrometer observations. For every coincident stratospheric observation, the NO_2(str) value and its uncertainty were calculated by integrating the vertical profiles of observed NO_2 concentrations and their errors, respectively, for altitudes between 18 and 50 km. For both SAGE II and HALOE data, integrations including altitudes below 18 km sometimes led to NO_2(str) values that were larger than NO_2(tot), especially for Zhigansk, which is characterized by a lower pollution level than Moshiri and Issyk-Kul. At lower altitudes, the relative errors in the SAGE II and HALOE NO_2 concentration data increase and exceed 100% below 18 and 14 km, respectively, even in the summer when stratospheric NO_2 abundances are largest. This suggests that the discrepancy could be accounted for by the SAGE II and HALOE NO_2 concentration profile errors. The summertime NO_2 amounts between the tropopause and 18 km were calculated assuming that the NO_2 concentration decreased monotonically from 18 km to the tropopause, where the concentration was set to zero. The calculated NO_2 amounts (~4, ~4, and $\sim 10 \times 10^{14}$ cm^{-2}, for Moshiri, Issyk-Kul and Zhigansk, respectively) are considered as the maximum uncertainty in NO_2(str) due to exclusion of NO_2 between the tropopause and 18 km.

[7] For each ground-based measurement site at SR and SS, the above calculations give two sets of NO_2(str) stratospheric column values, one from SAGE II and one from HALOE. To check consistency between these two sets, we used the criteria of a 2-hour difference and a 500-km distance between the SAGE II and HALOE observations to identify coincident SAGE II- and HALOE-based column data within 2° latitude of the ground-based measurement locations. Comparisons between the two sets of NO_2(str) columns showed that the median values of the differences (SAGE II minus HALOE) (±67% range) were 0.0 (+3.3/−2.7) × 10^{14} and 6.6 (+2.2/−5.0) × 10^{14} cm^{-2} for local SR and SS, respectively. For local SS, the median of the relative differences was 20% (+13%/−17%). Since the NO_2 data obtained by SAGE II at SS could be biased [*Bracher et al.*, 2005], the SAGE II NO_2(str) stratospheric column values at SS were adjusted to agree with the HALOE values at SS. The tropospheric NO_2 columns at SR or SS were then estimated by subtracting the SAGE II- or HALOE-based NO_2(str) values from the corresponding NO_2(tot) values. For example, the estimated NO_2(str) and NO_2(tot) values for Issyk-Kul are shown in Figure 1. In most of the cases over

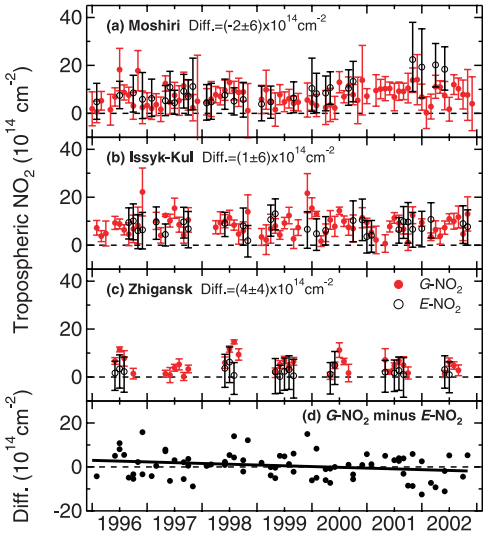

Figure 2. Time series of G-NO_2 (red) and E-NO_2 (black) values for (a) Moshiri, (b) Issyk-Kul, and (c) Zhigansk. For each site, the mean difference (G-NO_2 minus E-NO_2) (±standard deviation) is also shown. (d) The differences between G-NO_2 and E-NO_2 are all shown. The line is the linear least-square fit to the data.

the three sites, NO_2(tot) is greater than NO_2(str), indicating some contribution of tropospheric NO_2 to NO_2(tot).

[8] To derive F values, we used a global tropospheric chemistry model, CHASER [*Sudo et al.*, 2002], with 32 vertical layers (surface to ~5 hPa) and T42 horizontal resolution (~2.8° latitude × ~2.8° longitude). CHASER includes 53 chemical species and 139 reactions, including gas, liquid, and heterogeneous reactions, and takes into account O_x-NO_x-HO_x-CH_4-CO chemistry and the oxidation of non-methane hydrocarbons. For each of the ground-based observation sites used in this study, we calculated daily F values (~0.5–0.6 on average) by dividing the modeled tropospheric NO_2 column at the times of the GOME measurements by that at SR or SS. One-σ standard deviations (s.d.) of the monthly-averaged F values (~13–24%) are assumed to represent the uncertainty in the daily F value. After multiplying the F values with the above-estimated tropospheric NO_2 column abundances for both SR and SS, the E-NO_2 values were estimated and averaged for each month, yielding monthly-mean E-NO_2 values for each of the ground-based observation sites.

4. Results and Discussion

[9] The comparisons between the monthly-mean G-NO_2 and E-NO_2 tropospheric column amounts over Moshiri, Issyk-Kul, and Zhigansk are shown in Figures 2a–2c, respectively. Error bars for the G-NO_2 values represent 1σ s.d. of the monthly-mean G-NO_2 values over a 2° (in latitude) × 2° (in longitude) region centered on the ground-based measurement sites. Error bars for E-NO_2 represent the uncertainties in NO_2(str) and F, as described above. As shown in the figures, most of the G-NO_2 values agree with the E-NO_2 values to within their combined error ranges. The differences are nearly independent of season, suggesting that the uncertainty in E-NO_2, due to the use of constant AMF values and the seasonal change in sensitivity of the UV/vis. spectrometer observations to the troposphere, is smaller than the errors shown in the figures. Over 1996–2002, the mean differences (G-NO_2 minus E-NO_2) (±1σ s.d.) were -2 (±6) × 10^{14}, 1 (±6) × 10^{14}, and 4 (±4) × 10^{14} cm^{-2}, for Moshiri, Issyk-Kul, and Zhigansk, respectively. Thus, since no significant difference in the G-NO_2/E-NO_2 comparison results was found among the three locations, we next averaged all the differences between G-NO_2 and E-NO_2 values for all the locations. The mean difference was estimated to be 0.6 × 10^{14} cm^{-2}, with a 1σ s.d. of 6 × 10^{14} cm^{-2}, suggesting that a G-NO_2 variation exceeding 6 × 10^{14} cm^{-2} can be interpreted as being statistically meaningful over East Asia. We note that the values of E-NO_2 could be affected systematically by the errors in AMF and the weak sensitivity of UV/vis. spectrometer observations to the troposphere. However, the result may suggest that the systematic bias in the retrieved G-NO_2 data is less than 6 × 10^{14} cm^{-2}, if one considers previous validation studies made by *Heland et al.* [2002] and *Martin et al.* [2004]. They estimated the differences between G-NO_2 and the tropospheric NO_2 columns derived from aircraft observations to be less than 7 × 10^{14} cm^{-2} over Austria in spring of 2001 [*Heland et al.*, 2002] and less than 6 × 10^{14} cm^{-2} on average over eastern Texas and the southeast U.S. in the summers of 1999 and 2000 [*Martin et al.*, 2004].

[10] These previous comparisons have been made under polluted conditions (tropospheric NO_2 columns reaching 60 × 10^{14} cm^{-2}), whereas relatively clean sites have been used in the present study, due to the lack of ground-based UV/vis. spectrometer observation at polluted sites in East Asia. In addition, the UV/vis. spectrometer observations in East Asia were available only at SR and SS. Observations at SR and SS are generally less sensitive to tropospheric NO_2 than those in daytime. Thus, a ground-based UV/vis. spectrometer observation in polluted regions in daytime is highly desirable over East Asia to perform precise validation of future observations by nadir-viewing satellite instruments.

[11] The magnitude of the additional errors in E-NO_2 depends mainly on season and is almost constant for the same seasons. Therefore, we can assess a possible long-term drift in the G-NO_2 data over years 1996–2002 by

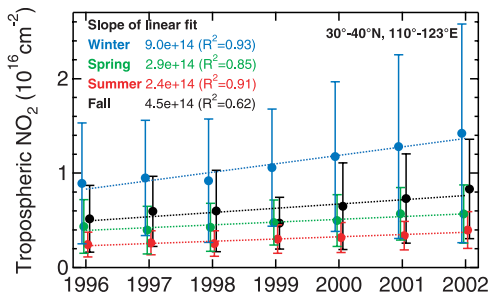

Figure 3. Time series of the mean G-NO_2 values in industrial areas of China (30°–40°N and 110°–123°E) for winter (December–February) (blue), spring (March–May) (green), summer (June–August) (red), and fall (September–November) (black). Error bars represent standard deviations of G-NO_2 over the region. The lines are the linear least-square fits to the data.

investigating the year-to-year variations in the differences between G-NO_2 and E-NO_2 for each season. Since the differences are nearly independent of season as described above, all the differences are plotted in Figure 2d. We find that the differences tend to decrease, at a rate of $\sim 0.7 \times 10^{14}$ cm^{-2} year^{-1} on average (Figure 2d). However, this trend is much smaller than the 1σ s.d. of all the differences estimated above and is negative, showing a counter trend towards the reported increasing trend in the industrial areas of China (30°–40°N and 110°–123°E) (Richter et al., submitted manuscript, 2005). Since insignificant, negative trends were seen in the time series of the G-NO_2/E-NO_2 differences at each site (not shown), the result is not affected by the different retrieval procedures employed at each site.

[12] We take into account the drift in G-NO_2 and estimate the linear trend in tropospheric NO_2 amounts over the industrial areas of China for each season (Figure 3). The most significant increasing trend in NO_2 occurred in wintertime ($\sim 9.0 \times 10^{14}$ cm^{-2} year^{-1}) with a correlation coefficient (R^2) of 0.93. As the observed trends are much greater than the estimated drift, these results are very similar to those of Richter et al. (submitted manuscript, 2005) and confirm their trend analysis. Accounting for the drift estimated above, the mean relative trends from 1996 to 2002 are estimated to be 8, 6, 8, and 7% year^{-1} for winter, spring, summer, and fall, respectively. Thus, a continuous increase in tropospheric NO_2 abundances likely occurred in the industrial regions of China between 1996 and 2002, at a rate of 7 ± 1% year^{-1}. This trend may be greater than that over the whole country, but is likely to represent the qualitative tendency over China. This additional information from GOME will be useful to reduce uncertainty in bottom-up emission estimates for China.

5. Conclusions

[13] To evaluate the long-term tropospheric NO_2 column data obtained by GOME (G-NO_2) in East Asia, we used tropospheric NO_2 abundances estimated based on observations by ground-based UV/vis. spectrometer and satellite-borne solar occultation sensors (SAGE II and HALOE) (E-NO_2) for three locations in Central and East Asia (Moshiri in Japan, Issyk-Kul in Kyrgyzstan, and Zhigansk in Russia). The mean differences between G-NO_2 and E-NO_2 were $(0.6 \pm 6.0) \times 10^{14}$ cm^{-2}, suggesting that there is no significant bias in the G-NO_2 data in East Asia. For making more precise comparisons, however, ground-based observations in polluted regions during daytime would be more suitable than those used in this study. The comparisons between G-NO_2 and E-NO_2 show that a slight drift in G-NO_2 data occurred, at an average rate of -0.7×10^{14} cm^{-2} year^{-1} from 1996 to 2002. However, this is much smaller than the observed increases in NO_2 over the industrial areas of China (30°–40°N and 110°–123°E), confirming the trend estimates made here and by Richter et al. (submitted manuscript, 2005). This multi-year G-NO_2 data showing a large increasing trend of 7 ± 1% year^{-1} over the region will be useful for reducing the uncertainty in bottom-up emission estimates for China.

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