Validation of POAM III NO$_2$ measurements

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[1]   We describe the Polar Ozone and Aerosol Measurement (POAM) III NO$_2$ measurements and associated errors and compare the POAM III data to correlative measurements obtained from satellite-, balloon-, and ground-based instruments. POAM III NO$_2$ densities are retrieved from 20 to 45 km, with a vertical resolution of about 1.5–2.5 km at altitudes below 40 km and increasing to more than 7 km at an altitude of 45 km. Predicted random errors are on the order of 5% in this altitude range. Sunspots and high aerosol extinction can cause errors in the NO$_2$ retrievals but generally affect only about 10% of the data or less, depending on the altitude. The agreement between POAM III NO$_2$ data and correlative observations is excellent, demonstrating that the POAM III measurements are reasonable in terms of their magnitude, profile structure, and temporal variations. The largest number of comparisons was made with the Halogen Occultation Experiment (HALOE) on the Upper Atmosphere Research Satellite. On average, POAM and HALOE agree to within about 0.2 ppbv from 20 to 33 km or within about 6% at most of these altitudes, with no systematic bias. Differences increase to about 0.7 ppbv (17%, POAM higher than HALOE) by 40 km. This difference decreases to about 12% after accounting for a recently discovered error in the HALOE retrievals. Differences decrease above 40 km and are slightly negative (0.1–0.2 ppbv on average) at 45 km, the top edge of the valid POAM III NO$_2$ altitude range. We conclude that the POAM III NO$_2$ profiles from 20 to 45 km are appropriate for scientific analysis and for the validation of NO$_2$ measurements from other instruments.   INDEX TERMS: 0394 Atmospheric Composition and Structure: Instruments and techniques; 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 3349 Meteorology and Atmospheric Dynamics: Polar meteorology

1.   Introduction

[2]   Polar Ozone and Aerosol Measurement (POAM) III [*Lucke et al.*, 1999] is a nine-channel (0.354–1.018 μm) solar occultation instrument designed to measure stratospheric profiles of ozone, nitrogen dioxide and water vapor densities, aerosol extinction at five wavelengths, and temperature. It was launched onboard the Satellite Pour l'Observation de la Terre (SPOT) 4 in March 1998 into a Sun synchronous orbit, and is currently operational. The instru-

ment is similar to its predecessor, POAM II [*Glaccum et al.*, 1996], with several design improvements described by *Lucke et al.* [1999]. Like POAM II, the latitude of the POAM III measurements varies slowly throughout the year between 55°N and 73°N and between 63°S and 88°S (Figure 1), with essentially identical coverage from year to year. All measurements in the Northern Hemisphere (NH) are made at satellite sunrise (sr), and all measurements in the Southern Hemisphere (SH) are made at satellite sunset (ss). This corresponds to local ss in the NH throughout the year, to local sr in the SH from early April to early September, and to local ss in the SH at other times of year.

[3]   Validation analyses for POAM III O$_3$ [*Lumpe et al.*, 2002a; C. E. Randall et al., Validation of POAM III O$_3$: Comparisons with ozonesonde and satellite data, submitted to *Journal of Geophysical Research*, 2002], water vapor [*Nedoluha et al.*, 2002], and aerosols [*Randall et al.*, 2001a] are presented elsewhere. These papers show that POAM III O$_3$, H$_2$O, and aerosols agree well with correlative measurements, and are valid for scientific use. Preliminary validation of the POAM III NO$_2$ data was presented by *Lucke et al.* [1999], and the NO$_2$ data have been used to show the

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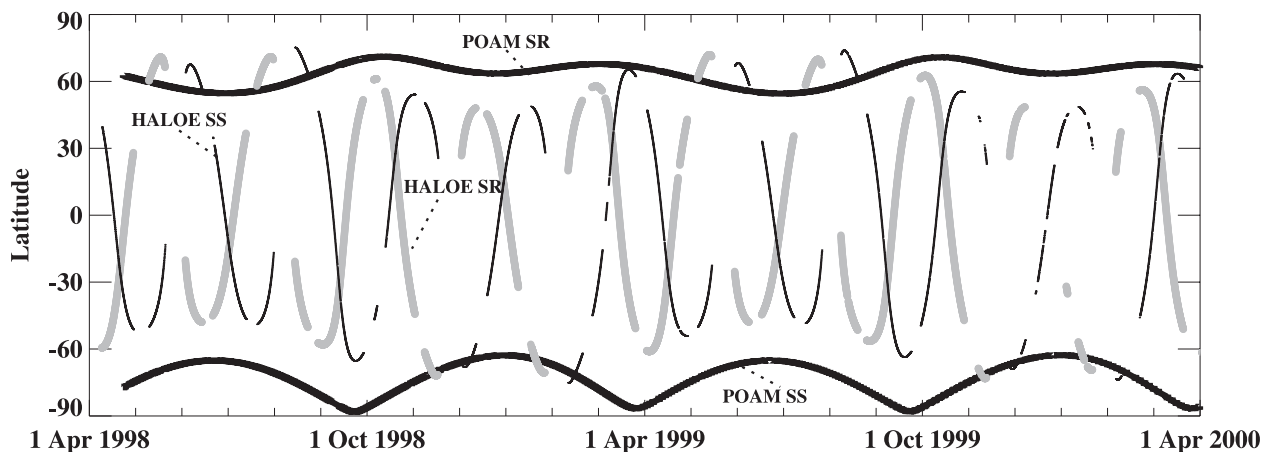
Figure 1. POAM III and HALOE measurement latitudes from April 1998 to March 2000. Satellite sunrise (sr) and sunset (ss) occultation measurements are indicated in the figure.

effects on the stratosphere of a large solar proton event that occurred in July 2000 [*Randall et al.*, 2001b]. Here we describe the POAM III version 3.0 NO$_2$ measurements in detail, their associated errors, and comparisons to correlative data.

2. POAM III NO$_2$ Data

[4] The POAM III version 3.0 retrieval algorithm is described by *Lumpe et al.* [2002b]. NO$_2$ is derived through differential measurements at 439.6 nm (NO$_2$-"on" channel) and 442.2 nm (NO$_2$-"off" channel). The fundamental quantity retrieved is NO$_2$ density; mixing ratios are calculated by dividing by the total atmospheric density obtained from the Met Office (UKMO) [*Swinbank and O'Neill*, 1994], interpolated in time and space to the POAM measurement locations. Figure 2 shows the theoretical random errors predicted by *Lumpe et al.* [2002b] for the POAM III NO$_2$ density retrievals. These predictions are the average relative errors based on simulations including mixing ratio profiles that spanned the range of conditions encountered by POAM, and are relatively insensitive to those conditions since the random errors tend to vary according to absolute
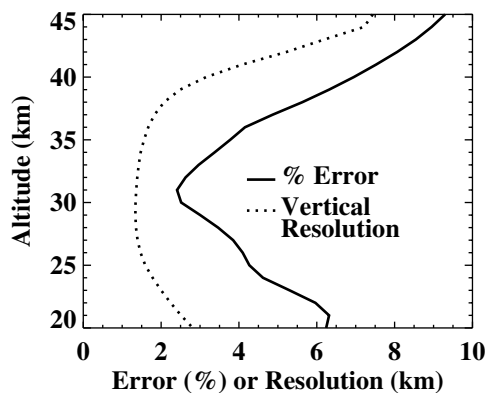
mixing ratios. The random error minimizes around 30 km at a value of 2.5%, increasing to ∼6.5% at 20 km and to ∼9.5% at 45 km. The vertical resolution of the retrieved NO$_2$, which is also shown in Figure 2, is about 1.5 km or better between altitudes of 25 and 35 km, increasing to nearly 3 km at altitudes of 20 and 40 km, and to more than 7 km at an altitude of 45 km.

[5] Sporadically, random errors in the POAM III NO$_2$ retrievals can arise during high aerosol loading conditions, or during strong sunspot activity *Lumpe et al.* [2002b]. Since the POAM NO$_2$ measurement is a differential measurement, the retrievals are to first order independent of the aerosol loading. In times of very high extinction, however, as in the presence of polar stratospheric clouds (PSCs), small errors in the aerosol/NO$_2$ separation can cause random artifacts in the NO$_2$ data. Figure 3 illustrates the effect on the NO$_2$ retrievals of high aerosol extinction. Here we plot the measured POAM III NO$_2$ concentrations in the SH at an altitude of 23 km, during 1999. We chose this altitude and
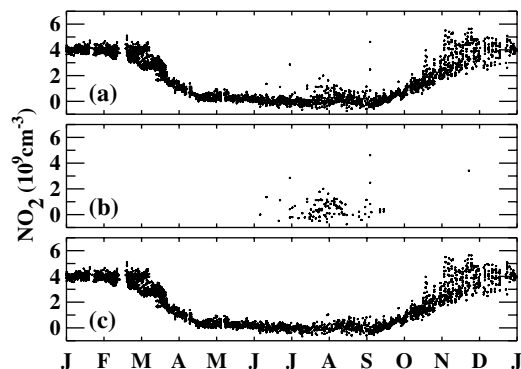

Figure 3. POAM III NO$_2$ concentrations at 23 km in the SH during 1999. (a) All NO$_2$ measurements except those that have been screened for possible sunspot artifacts (see Figure 4). (b) NO$_2$ measurements where the predicted error from aerosol interference exceeds the error threshold limit, leading to possible artifacts in the NO$_2$ retrieval. (c) Same as (a), but with the points in (b) removed.

Figure 2. Random error (solid) and vertical resolution (dotted) profiles for POAM III NO$_2$.

hemisphere since it represents a worst case in terms of the numbers of data points that are possibly affected by high extinction aerosols; other years are similar. In panel a we plot the NO_2 concentrations for all measurements in 1999, except those possibly affected by sunspots (see below). There are two time periods when the variability in the measurements is particularly high—July/August and November/December. During the latter time period, POAM III is sampling both inside and outside the vortex, and the observed NO_2 variability is due to geophysical variability at this time. During the July/August period, however, we believe that much of the observed variability is due to errors from incorrect NO_2/aerosol separation in the presence of PSCs. *Lumpe et al.* [2002b] have quantified the aerosol separation errors for all POAM measurements, at all altitudes. At each altitude below 31 km, using all of the POAM III measurements, we have determined the 3σ standard deviation from the average error calculated by *Lumpe et al.* [2002b], and have assigned this value as a threshold limit. All measurements for which the calculated error exceeds this threshold are flagged as having possible aerosol artifacts. In Figure 3b we plot only the NO_2 concentrations for the flagged data points. As expected, most of the flagged measurements occur during the July/August PSC season. In Figure 3c we plot the same measurements as in panel a, but omitting all of the flagged data shown in panel b. Note that the variability during the July/August time period is significantly less in this plot. In the SH, the flagged data points comprise about 12% of the entire data set from 20 to 23 km, about 8% at 25 km, and less than 1% at 30 km (mostly during the SH winter). In the NH, fewer than 0.5% of the data at any altitude are flagged as having possible aerosol artifacts.

[6] The sunspot errors described by *Lumpe et al.* [2002b] arise primarily from errors in the POAM III normalization procedure. Sunspots cause an attenuation of the observed solar intensity, I_o, as the POAM instrument scans across the sunspot's location on the solar disk during the sunscan (the sunscan is used as the I_o signal to which the atmospheric attenuation is normalized; see the work of *Lumpe et al.* [2002b]). In principle, the sunspot attenuation would be accounted for during the normalization procedure. However, improper registration of the I and I_o signals due to small uncertainties in our knowledge of the POAM pointing relative to the sunspot location can lead to normalization errors, and thus to random artifacts in the NO_2 retrievals [*Lumpe et al.*, 2002b]. We screen the POAM III NO_2 measurements for possible sunspot artifacts in a manner analogous to the aerosol error screening. That is, we flag all measurements for which the calculated sunspot error exceeds the average sunspot error at each altitude by some threshold value; we have empirically defined this value as the 2σ variation from the average error.

[7] Figure 4a shows the observed POAM III NO_2 concentrations in the SH during 2000 at 35 km. This is one of the altitudes most affected by sunspots, and 2000 was a year of very high solar activity; there is not a significant difference between the NH and SH in terms of sunspot effects. There are a number of periods that last from about a day to a week where there are significant deviations from the average NO_2 measurements at that time. Figure 4b shows only those measurements that were flagged, as described above, as
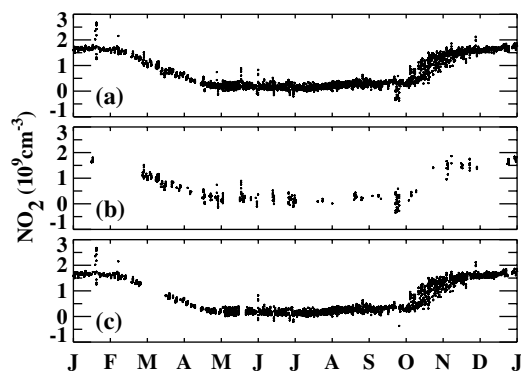

Figure 4. POAM III NO_2 concentrations at 35 km in the SH during 2000. (a) All NO_2 measurements except those that have been screened for possible aerosol artifacts (see Figure 3). (b) NO_2 measurements where the predicted sunspot error exceeds the error threshold limit, leading to possible artifacts in the NO_2 retrieval. (c) Same as (a), but with the points in (b) removed.

possibly having errors due to sunspots. Flagged data constitute about 10% of the measurements from 20 to 45 km. Figure 4c shows the same data as Figure 4a, but excluding the flagged points in Figure 4b. Many of the outlying points in Figure 4a are no longer evident in Figure 4c. It is also apparent, however, that a number of points are flagged that in fact appear to align well with the rest of the data (indicating that perhaps the sunspot did not cause an error of the magnitude predicted), and that there are also a number of points that were not flagged but perhaps should have been. For instance, the measurements on 17–18 January 2000 deviate significantly from those around them. At this time, attenuation due to relatively small sunspots was present in the POAM sunscans, but the magnitude of the attenuation was such that the predicted error was not outside the threshold limit. That the sunspot screening is not perfect is well understood [*Lumpe et al.*, 2002b]: the actual errors introduced in the POAM NO_2 retrievals depend on our uncertainty in the position of the sunspot on the Sun; if the assumed sunspot position is accurate, then even a very large sunspot will not cause an error in the measurement. On the other hand, if the assumed position is incorrect, then even a very small sunspot can cause an error. The comparisons shown in the rest of the paper exclude any events for which the NO_2 data are flagged as possibly having a sunspot artifact, as described here. We are confident that our conclusions are not biased by any false negative flag determinations (i.e., by the inclusion of data affected by sunspots). POAM data users should be aware, however, that sunspots can cause random artifacts in the NO_2 retrievals of the type seen in Figure 4, and should be cautious when interpreting individual NO_2 profiles in the presence of sunspots.

[8] The only known source of bias in the POAM NO_2 retrieval is related to the treatment of diurnal variations along the line of sight. It is well known that because of rapid variations near sunrise and sunset, NO_2 exhibits strong variations along a solar occultation measurement line of sight [e.g., *Newchurch et al.*, 1996]. As discussed by *Lumpe et al.* [2002b] the POAM III retrievals do not include corrections

for such variations, because the magnitude of the correction is strongly dependent on the photochemical model employed in the retrieval algorithm. Neglecting the line of sight variations makes little difference above about 25 km, but depending on the location and season, *Newchurch et al.* [1996] estimate that neglecting the diurnal variations can result in systematic overestimates in NO$_2$ of up to about 20% at 20 km. There are no other known sources of systematic error in the NO$_2$ retrievals. However, the NO$_2$ cross sections used in the POAM III retrievals, from the work of *Harder et al.* [1997], have quoted uncertainties of ±4%. A cross-section error of this magnitude, if systematic (not random), would translate directly to a 4% bias in the retrieved NO$_2$ densities. Also, *Randall et al.* [2001a] suggested that some of the discrepancies between the POAM III and Stratospheric Aerosol and Gas Experiment (SAGE) II aerosol measurements could be interpreted as arising from a very small timing error in the POAM III measurements, equivalent to an altitude error of less than 175 meters. The effects of such an error on the NO$_2$ retrievals will be discussed below.

3. Correlative Comparisons

3.1. Correlative NO$_2$ Data

[9] The two satellite instruments that offer the most opportunities for correlative measurements with POAM III NO$_2$ profiles are SAGE II and the Halogen Occultation Experiment (HALOE). Of these, only the HALOE data are used in this paper since, at the time of this writing, the newest publicly released version (6.0) SAGE II NO$_2$ measurements differ substantially from other correlative observations, and an even more recent version (6.1) has yet to be validated. HALOE uses the solar occultation technique in the mid infrared spectral region to measure NO$_2$ as well as other gas and aerosol profiles. HALOE occultation events occur primarily at mid latitudes, but overlap the POAM latitudes several times a year for several days at a time (Figure 1), and thus are excellent for correlative observations. All results here use version 19 HALOE data.

[10] The version 17 HALOE NO$_2$ data were validated by *Gordley et al.* [1996], who concluded that between 25 and 35 km, the HALOE NO$_2$ was accurate (agreed with correlative measurements) to about the ±10% level, and exhibited essentially no bias in this altitude region. Below 25 km, the HALOE NO$_2$ exhibited a low bias of up to about 0.5 ppbv; the bias was less when NO$_2$ concentrations were large and the aerosol extinction was low. Between 35 and 40 km the comparison results were variable, but indicated a possible low bias of 5%. The quality of the version 18 HALOE NO$_2$ data is characterized on the HALOE web page (http://haloedata.larc.nasa.gov). The NO$_2$ data is described as excellent from the tropopause to 25 km in clear air conditions, but as exhibiting a low bias in the presence of aerosols—this should not be significant for the comparisons presented here, since they pertain to the background aerosol conditions of 1998–2000 [e.g., *Randall et al.*, 2001a]. Version 18 data from 25 to 45 km agree with correlative observations to the ±10–15% level, with no obvious bias (the error in version 17 leading to the low bias between 35 and 40 km, which was related to a problem in the pressure retrieval, was fixed in version 18). The primary differences between version 18 and
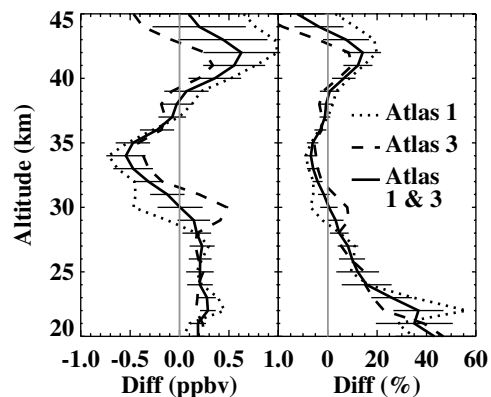

Figure 5. Average absolute (left) and relative (right) differences between HALOE version 19 and ATMOS version 3.0 NO$_2$ profiles (ATMOS−HALOE) for the coincidences noted in Table 1. "Error" bars represent the 1σ uncertainties in the mean differences for the combined Atlas 1 and Atlas 3 case.

version 19 HALOE NO$_2$ data were seen in the altitude region below about 20 km, and are not relevant for the comparisons presented here.

[11] Because of the lack of published validation studies for version 19 HALOE NO$_2$ measurements, we have compared them to data from the Atmospheric Trace Molecule Spectroscopy Experiment (ATMOS) [*Farmer*, 1987; *Gunson et al.*, 1996] to give further confidence in the appropriateness of using these profiles as correlative measurements for the POAM III NO$_2$ validation. ATMOS is a shuttle-based solar occultation Fourier transform infrared (FTIR) spectrometer used for remotely measuring the profiles of a number of different trace gas species from the troposphere to the mesosphere. We use ATMOS version 3.0 data, which are described on http://remus.jpl.nasa.gov/atmos (F. W. Irion et al., in preparation, 2001). The HALOE retrievals include a correction for diurnal variations along the line of sight, so we compare them to ATMOS data that also include this correction. The uncorrected version 2.0 ATMOS NO$_2$ data have an estimated accuracy of about 6% in the 20–45 km range used here, based on uncertainties in the spectroscopic parameters and the ATMOS pressure determination [*Abrams et al.*, 1996]. Because of uncertainties in ozone that feed into the diurnal correction, the error in the diurnally corrected NO$_2$ near 20 km is about 9% or greater. The version 3.0 retrievals have similar accuracy to version 2.0 NO$_2$ from 20 to 45 km, and random errors that increase from about 10% at 30 km to at least 35% at 20 km (F. W. Irion, personal communication, 2001).

[12] Figure 5 shows the average differences between coincident HALOE and ATMOS measurements (defined to occur within ±4° in latitude, ±12° in longitude, and ±2 hours in time) from the Atlas space shuttle flights 1 and 3, which took place during 1992 and 1994 (see Table 1). There were no measurements that fit the above coincidence criteria during the Atlas 2 flight. Although there were a total of only 14 coincident profiles included in these comparisons, the results are consistent with the information noted above. The HALOE version 19 NO$_2$ data are on average only about 0.2–0.3 ppbv lower than ATMOS below 29 km. Because

Table 1. HALOE/ATMOS Coincidences during Atlas Flights 1 and 3[a]

Date	Type	Events	Latitude	Distance, km	ΔLatitude, deg	ΔLongitude, deg	ΔTime, min
At 1: 27–29 March 1992	ss/ss	7	−45.8	586 ± 201	−0.2 ± 3.0	−2.8 ± 6.6	0 ± 1
At 3: 7–8 November 1994	ss/ss	7	34.1	618 ± 321	−0.1 ± 2.4	−1.5 ± 7.1	0 ± 4

[a]Columns: (1) inclusive dates of the coincident events, (2) type of occultation event from the perspective of the respective satellites (ATMOS/HALOE: sunrise (sr), sunset(ss)), (3) number of events included in the comparisons, (4) average HALOE latitude, and average difference (ATMOS – HALOE) in (5) distance, (6) latitude, (7) longitude, and (8) time. All coincidences occurred at local sunset.

NO$_2$ mixing ratios are so low below 25 km, this translates into relative (%) differences of up to about 40%. Above 25 km the two instruments compare to within about ±10%. In the context of these comparisons, it is important to note that the ATMOS v3.0 retrievals use spectral parameters from the 1996 HITRAN compilation. Because of anticipated future changes in these parameters, it was decided that the HALOE retrievals would continue to use the 1992 HITRAN spectral parameters. HALOE NO$_2$ would be reduced by about 10% if the 1996 HITRAN parameters were used, and the ATMOS – HALOE comparisons in Figure 5 would change accordingly. It should be further noted, however, that the HITRAN 2000 compilation includes a major revision to the NO$_2$ line parameters (www.hitran.com), and it is currently unclear how these changes will affect the HALOE (or ATMOS) retrievals.

[13] In addition to the statistical analysis of POAM– HALOE comparisons, we present comparisons to small numbers of NO$_2$ profiles measured by the MkIV Interferometer [*Toon*, 1991], the Laboratoire de Physique Molèculaire et Applications and Differential Optical Absorption Spectroscopy (LPMA/DOAS) balloon gondola instrument [*Camy-Peyret et al.*, 1993; *Ferlemann et al.*, 2000], and the Système D'Analyze par Observations Zénithales (SAOZ) balloon instrument. The MkIV and LPMA/DOAS correlative measurements were made during the recent SAGE III Ozone Loss and Validation Experiment (SOLVE)/Third European Stratospheric Experiment on Ozone (THESEO) 2000 campaign. The SAOZ measurements were made prior to and during the SOLVE/THESEO 2000 campaign. Finally, we also show comparisons between vertically integrated POAM NO$_2$ and ground-based column NO$_2$ measurements from 1998 to 2000 made by New Zealand's National Institute of Water and Atmospheric Research (NIWA).

3.2. Statistical Analysis

[14] In this section we describe the statistical calculations used to compare POAM to the HALOE data. Similar analysis procedures were performed for the comparisons between POAM and the other correlative data sets, where multiple measurements were available. Since there are many fewer profiles contained in the other correlative data sets, however, these other comparisons provide less conclusive results, and are shown primarily as a confidence check on the POAM–HALOE results.

[15] The HALOE and POAM III data sets were searched for spatially and temporally coincident events, which are listed in Table 2. Coincidences were determined by requiring correlative measurements to occur within ±4° in latitude, ±12° in longitude, and ±2 hours in time of the POAM III measurements. As noted in the table, the actual differences were usually substantially smaller than the requirements, with average distances between the POAM III and HALOE measurements of around 450 km in the NH and 350 km in the SH. Temporal differences are generally larger in the SH; however, since all measurements occur at local sunrise or local sunset for both the HALOE and POAM data sets, these time differences primarily reflect longitudinal differences, and are not expected to be significant in terms of true diurnal variations in NO$_2$ [e.g., *Randall et al.*, 1998, and references therein]. Overall, the small hemispheric biases seen in Table 2 for the latitude, longitude and time differences are insignificant with regard to the comparisons presented here, because the geophysical variations in NO$_2$ expected from such spatial and temporal differences are smaller than the combined uncertainties in the measurements themselves. There is a bias in the occultation event types (sr or ss from the perspective of the satellite) of the coincidences, in that all NH coincidences occurred with HALOE satellite ss events and POAM satellite sr events, whereas in the SH, the coincidences contained only POAM satellite ss events, but both satellite sr and satellite ss events from HALOE. This could affect the comparisons if either HALOE or POAM exhibit a bias in their NO$_2$ retrieved from satellite sr occultations compared to satellite ss occultations. *Gordley et al.* [1996] showed that for high-latitude

Table 2. Catalog of POAM III/HALOE Coincidences[a]

Time period	Type	Events	Latitude	Distance, km	ΔLatitude, deg	ΔLongitude, deg	ΔTime, min
(a) 11–12 June 1998	sr/ss	21	55.4	493 ± 136	−2.6 ± 0.9	−0.9 ± 6.8	−18 ± 7
(b) 20–21 August 1998	sr/ss	18	61.9	419 ± 178	−2.5 ± 0.9	0.6 ± 7.2	−11 ± 4
(c) 16–24 March 1999	sr/ss	102	67.0	412 ± 118	2.6 ± 0.7	0.1 ± 6.9	0 ± 1
(d) 16–17 August 1999	sr/ss	14	60.6	424 ± 190	−1.8 ± 0.9	1.4 ± 7.9	−8 ± 4
(e) 14–15 March 2000	sr/ss	22	67.4	535 ± 77	3.9 ± 0.0	1.3 ± 7.0	−3 ± 0
(f) 13–14 November 1998	ss/sr	14	−70.0	417 ± 48	1.8 ± 0.1	5.2 ± 9.2	−88 ± 17
(g) 2–8 December 1998	ss/ss	82	−65.0	369 ± 146	0.2 ± 2.1	0.3 ± 6.9	−20 ± 52
(h) 9–14 February 1999	ss/ss	61	−71.3	330 ± 122	0.3 ± 2.4	−0.9 ± 6.7	−10 ± 36
(i) 9–10 November 1999	ss/sr	11	−71.3	248 ± 32	1.8 ± 0.0	4.4 ± 1.4	−91 ± 14
(j) 26–30 November 1999	ss/ss	15	−66.3	383 ± 120	1.8 ± 0.7	−0.7 ± 8.2	−60 ± 35

[a]Columns: (1) inclusive dates of the coincident events, (2) type of occultation event from the perspective of the respective satellites (POAM/HALOE: sunrise (sr), sunset(ss)), (3) number of events included in the comparisons, (4) average POAM latitude, and average difference (POAM – HALOE) in (5) distance, (6) latitude, (7) longitude, and (8) time. All coincidences occurred at local sunset.

spatially coincident HALOE observations that occurred during successive sunsets and sunrises, the NO$_x$ (NO + NO$_2$) measured at sunrise was lower than the values predicted based on the sunset measurements by up to about 2 ppbv above 10 mbar. However, as shown below, we see no systematic difference between the POAM−HALOE comparisons made with HALOE ss and HALOE sr events. We discuss possible POAM sr/ss biases when evaluating the differences between the NH and SH comparisons.

[16] Difference profiles between POAM III and the HALOE measurements were calculated using equation (1):

$$\Delta_i = \frac{100}{n_i} \sum_{n_i} \left[\frac{(POAM_i - Corr_i)}{Avg_i} \right] \qquad (1)$$

Here $POAM_i$ and $Corr_i$ refer to the POAM III and correlative (HALOE) measurements, respectively, at the ith altitude. Avg_i refers to the average of $POAM_i$ and the correlative (HALOE) data for calculations of the relative (%) differences, and $Avg_i = 1.0$ for the absolute differences. The number of coincident events at each altitude, n_i, varies due to points flagged as missing or suspect. For both POAM and the correlative data, this of course includes those events/altitudes for which the data files were populated with fill values. For POAM, this also includes points flagged for aerosol or sunspot errors, as described above. For HALOE, this includes all data where the quoted error was larger than 100%. We calculate the standard deviation of the distribution (or the sample variance, σ) to quantify the spread in the individual measurement difference pairs as:

$$\sigma = \sqrt{\frac{1}{N-1} \sum (x_i - \bar{x})^2} \qquad (2)$$

where x_i refers to the difference between POAM and the correlative measurement, $\bar{x}$ is the average of all differences, and N is the number of event pairs. All "error" bars on the differences reported below (not just the POAM−HALOE comparisons) refer to the uncertainty in the mean difference, or $\sigma/\sqrt{N}$.

4. Results

4.1. HALOE Comparisons

[17] Figure 6 shows the evolution of POAM NO$_2$ mixing ratios at 30 km from May 1998 to May 2000. In both hemispheres, mixing ratios are larger in summer than in winter, because of sequestration of NO$_2$ in the polar N$_2$O$_5$ (and HNO$_3$) reservoir during the winter, and the effects of the seasonally varying NO$_2$/NO equilibrium [*Randall et al.*, 1998]. Superimposed are the coincident HALOE measurements, as defined above and in Table 2. Qualitatively, the POAM and HALOE NO$_2$ mixing ratios are in agreement in all coincident time periods. Not only do they agree in magnitude, but both instruments show the same temporal changes in NO$_2$ variability, with small variances in nonvortex comparison periods (June and August in the NH, February in the SH) and larger variances when the polar vortex was present (March in the NH, November and December in the SH). The increased variation during comparison periods when the vortex is present results from measurements being made both inside and outside the vortex. For example, we
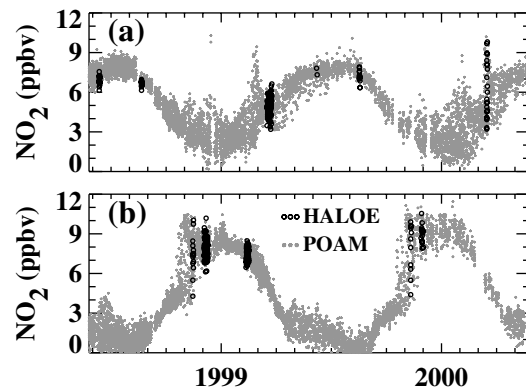

Figure 6. Temporal variation in NO$_2$ mixing ratios at 30 km measured by POAM III (gray points) from May 1998 to May 2000 in the NH (a) and SH (b). Coincident HALOE measurements are overplotted (black circles).

overplot the longitudinal variation in the POAM and HALOE NO$_2$ mixing ratios at 800 K (about 30 km) in Figure 7, for March in the NH (coincident time periods c and e) and November in the SH (coincidence time periods f and i). Superimposed are the potential vorticity values at the POAM measurement locations. In all cases, the higher NO$_2$ mixing ratios correspond to measurement locations outside the polar vortex (low potential vorticity magnitudes). This is due primarily to two effects: descent of relatively NO$_x$-poor air in the vortex leading to lower mixing ratios inside the vortex than outside, and poleward transport of relatively NO$_x$-rich air from lower latitudes, leading to higher NO$_2$ mixing ratios outside the vortex. Although not shown, similar behavior is seen during other time periods when the mixing ratios consistently exhibit larger variations than average (e.g., in the NH in late February 1999 and late January/early February 2000).

[18] The average profiles measured by POAM and HALOE for the coincident events in the individual time periods in Table 2 are shown in Figure 8. The agreement is remarkably good, with differences that are in most cases statistically insignificant. Nevertheless, there are consistent, systematic differences between about 35 and 42 km, where POAM is higher than HALOE by up to about 1 ppbv. This is discussed further below. Figure 8 also shows the standard deviations of the distributions, as defined in equation (2). As expected from the time series shown above (Figure 6), the variations measured by POAM and HALOE agree quite well within each time period. These plots indicate that the magnitude of the variations is relatively constant between about 25 and 40 km, with increasing (and occasionally divergent) variations near 20 and 45 km. These altitudes are at the edges of the valid range for the retrievals, and correspond to altitudes with very low NO$_2$ mixing ratios, so we attribute the increased variations to increased uncertainty in the retrievals. The most notable disagreement between the POAM and HALOE variances is near 25 km in panel e. At this altitude and time, mid-March 2000, the vortex was rapidly varying, and many of the POAM measurements were located very close to the vortex edge. Thus, not only were the variations larger in this time period than in other coincidence time periods, but the nearly 4° latitude difference between
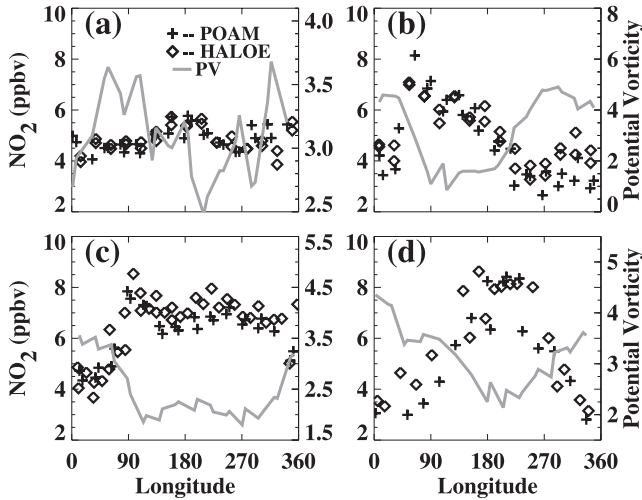
Figure 7. Longitudinal variations in NO$_2$ at 800 K (~30 km) measured by POAM and HALOE for the (a) 19–20 March 1999 and (b) 14–15 March 2000 coincidence time periods in the NH and for the (c) 13–14 November 1998 and (d) 9–10 November 1999 coincidence time periods in the SH. All POAM and HALOE measurements in these time periods are included, not simply the coincident events. The solid gray lines correspond to the potential vorticity (PV, 10^{-4} m^2 kg^{-1} s^{-1} K) at the POAM measurement locations (the PV was multiplied by −1 in (c) and (d)).

HALOE and POAM resulted in larger differences between the HALOE and POAM observed variances. In order to avoid these types of effects, one can use equivalent latitude rather than geographic coordinate coincidence criteria. However, since the total number of POAM/HALOE coincidences is so large, the geophysical variations are insignificant with regard to our overall conclusions regarding the validity of the POAM NO$_2$ data. As shown in Figure 8 and below, the POAM–HALOE comparisons in vortex and nonvortex periods are similar in character. For the other correlative measurements, we note specifically those cases where equivalent latitude considerations are relevant.

[19] Figure 9 shows the results of the statistical comparisons between POAM and HALOE for all of the individual coincidence time periods listed in Table 2. At most altitudes, in most time periods, and in both hemispheres, the two instruments agree very well, with average differences less than 0.5 ppbv. For the two cases with the smallest uncertainties in the mean (panels c and g), the average differences below 35 km are less than ~0.2 ppbv. We also note that the SH comparisons with HALOE sr events (periods f and i) are similar in character to the SH comparisons with HALOE ss events (periods g, h, and j); this is the origin of the statement made in section 3.2 that there does not appear to be any effect of a HALOE sr/ss bias in the comparisons shown here. As mentioned earlier, however, POAM NO$_2$ mixing ratios are consistently higher than HALOE between 35 and 42 km. Additionally, because of the low NO$_2$ mixing ratios near 20 and 45 km, relative differences at these altitudes can reach 50% or greater.

[20] Figure 10 shows the overall average POAM–HALOE differences in each hemisphere. Between 22 and 33 km the observed differences are remarkably small, and suggest that the POAM data are of excellent quality.
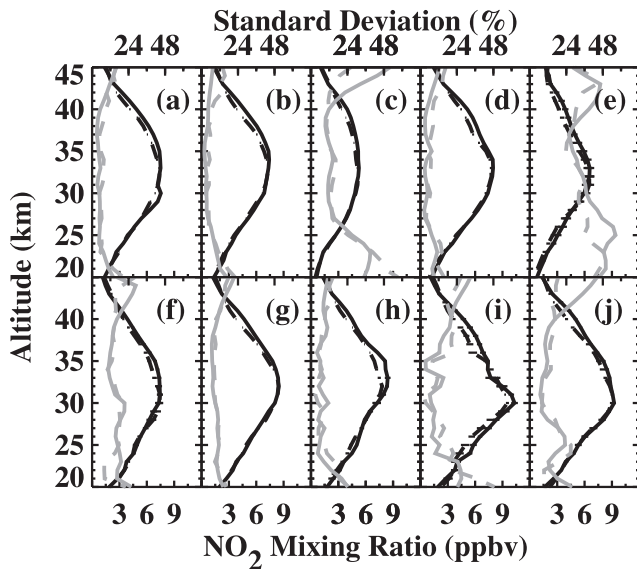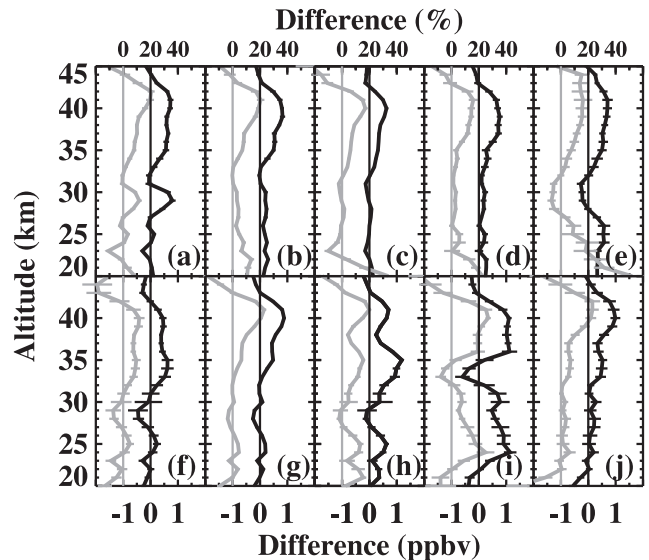

Figure 8. Average POAM III (black solid) and HALOE (black dot-dashed) NO$_2$ mixing ratio profiles (bottom axis) in the NH (top) and SH (bottom) for the corresponding coincidence time periods listed in Table 2. "Error" bars, which are often smaller than the width of the profile lines themselves, represent the 1σ uncertainty in the mean profiles, as described in the text. Also plotted are the standard deviations (top axis) of the distributions for POAM III (gray solid) and HALOE (gray dashed) as defined in equation (2).

Figure 9. Average POAM−HALOE absolute differences (black lines, bottom axis) in the NH (top) and SH (bottom) for the corresponding coincidence time periods listed in Table 2 (see equation (1)). Also plotted are the relative (%) differences (gray lines, top axis). "Error" bars represent the 1σ uncertainties in the mean profiles.
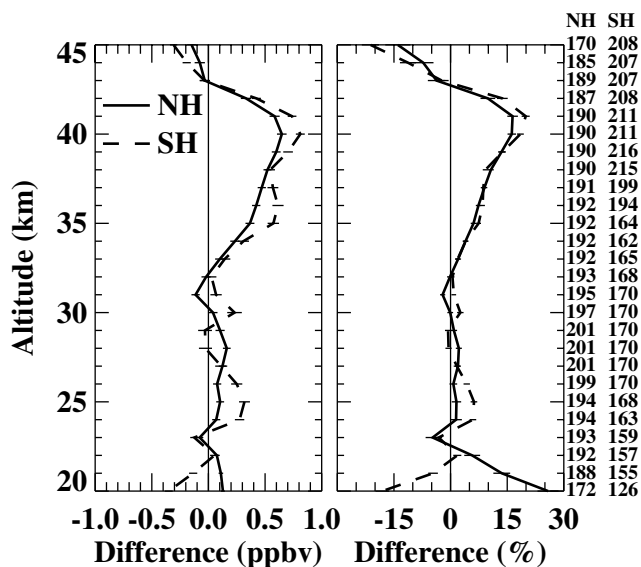
Figure 10. Average of all POAM−HALOE differences in the NH (solid) and SH (dashed) in absolute (left, ppbv) and relative (right, %) units. The numbers of events included in the comparisons at each altitude are given next to the right vertical axis. "Error" bars represent the 1σ uncertainties in the mean differences.

Furthermore, we note that at all altitudes except near 20 km (see below), there is not a significant bias between the NH and SH comparisons. This is particularly important since the NH POAM occultations all occur as spacecraft sr events, whereas all SH POAM occultations occur as spacecraft ss events. That there is not a substantial difference in the NH and SH comparisons suggests that any sr/ss bias in the POAM measurements [see *Randall et al.*, 2001a] is not significant for the NO₂ retrievals. The two most prominent features in Figure 10 that warrant further discussion are the positive differences between about 35 and 42 km, and the hemispheric bias in the relative differences at 20 km.

[21] Errors in NO₂ retrievals from both instruments are expected to increase as NO₂ decreases, so it is not surprising that the differences between the two measurements would increase above 35 km and below 22 km. As mentioned above, previous comparisons between HALOE and other correlative measurements show differences at the ±10−15% level. The maximum POAM−HALOE differences of about 17% near 40 km are only slightly outside this range, but are clearly systematic, not random in nature. The ATMOS− HALOE differences shown in Figure 5 hint at a possible low HALOE bias between 40 and 42 km, but this is only marginally significant; and if anything, Figure 5 would imply a high HALOE bias near 35 km. We noted above that there is some uncertainty in the NO₂ line parameters being used in the HALOE retrievals. Spectroscopic errors would result in a systematic bias in the HALOE data. However, because of the weak line limit condition in the HALOE band pass, verified by retrieval sensitivity studies, spectroscopic errors would not account for the altitude dependence seen in the POAM−HALOE differences. Nevertheless, prior to the availability of the POAM NO₂ data, there has not been an opportunity for such numerous

correlative comparisons with HALOE. Thus, we have carefully considered other possible error sources in the HALOE retrieval algorithm that might lead to a low bias, and have identified one such error. Aerosol extinction, while very low near 40 km, contributes significantly to the HALOE NO₂ channel because of the optically thin NO₂ signal at this altitude and wavelength. Recent simulations have shown that due to a high bias in the HALOE aerosol retrieval [*Hervig et al.*, 1996], compounded by the extrapolation of aerosol extinction to altitudes above the maximum aerosol retrieval altitude, the HALOE algorithm systematically overestimates the aerosol contribution to the NO₂ channel above 35 km. This leads to a systematic underestimate in retrieved NO₂, with a magnitude of 3−7% near 40 km, depending on the NO₂ mixing ratio. This error will be corrected in the next version of the HALOE retrievals, but at this time accounts for about 30% of the observed POAM− HALOE difference near 40 km. Tests for similar effects in the POAM data showed that any error in the retrieved NO₂ would be less than 1% near 40 km, because the aerosol contribution to the differential NO₂ measurement is significantly smaller than the NO₂ contribution itself. We have also considered other possible error sources in the HALOE retrievals that might lead to a low bias near 40 km, but have not conclusively identified any.

[22] To investigate the possibility that a systematic high bias in the POAM data contributes to the differences between 35 and 42 km, we have modeled the two most likely error sources: the decreasing altitude resolution of the POAM measurements (Figure 2), and the increasing contribution of the a priori to the POAM retrievals, as altitude increases. We have determined that these effects are consistent with the small negative POAM−HALOE differences near 45 km, but not with the observed differences between 35 and 42 km. At altitudes lower than 41 km, for instance, the POAM altitude resolution is close to the 2.3 km HALOE altitude resolution, and the dependence on the a priori is negligible [*Lumpe et al.*, 2002b]. Furthermore, the a priori NO₂ profile is on average lower than the POAM measurements, so even if it were contributing to the POAM retrieval, it would shift the POAM data to lower values, and thus would not result in a positive POAM bias. Finally, we have also performed the POAM−HALOE comparisons using both mixing ratios and concentrations, and on both altitude and pressure scales, with similar results, indicating that conversions between these quantities are not responsible for the differences. Therefore, at this time we attribute roughly one third of the POAM−HALOE NO₂ difference from 35 to 42 km to a low bias in the HALOE data, leaving a maximum residual difference of about 12% near 40 km that is not currently understood.

[23] The average absolute difference between POAM and HALOE in the NH at 20 km is less than 0.2 ppbv, but corresponds to a relative difference of 30%. As mentioned above, the POAM retrievals do not correct for diurnal variations in NO₂ along the line of sight. The HALOE retrievals do make this correction, however, and thus HALOE NO₂ mixing ratios are expected to be lower than the POAM retrievals below about 25 km [*Newchurch et al.*, 1996]. Because the diurnal variations along the line of sight are caused by photochemical reactions, and because there is a large seasonal variation in NO₂ mixing ratios, the magni-

Table 3. MkIV/POAM Coincidences[a]

Date	MkIV Latitude, deg	POAM Latitude, deg	MkIV Longitude, deg	POAM Longitude, deg	MkIV Time, hours	POAM Time, hours
3 December 1999	65.1	64.6	19.2	9.1	13.4	13.9
		64.6		34.4		13.9
15 March 2000	68.8	67.3	33.0	39.3	6.70	17.8

[a]Columns: (1) coincidence date, (2) average MkIV latitude, (3) POAM latitude, (4) average MkIV longitude, (5) POAM longitude, (6) average MkIV local time, and (7) POAM local time.

tude of the line of sight correction, in both an absolute and a relative sense, depends strongly on season and latitude. Therefore, we have run a modified HALOE retrieval algorithm on events in the time periods most significant for the POAM–HALOE comparisons (c, g, and h in Table 2). In this algorithm, the diurnal correction routine was turned off, so in that respect the HALOE data were retrieved in a manner analogous to the POAM data. As expected, the correction was very small (less than 5%) above about 20 km for the summertime SH events in December 1998 and February 1999. However, for the events near the spring equinox in the NH in March 1999, the correction gradually increased below 30 km to about 30% (mixing ratios higher without the correction) near 20 km. These results suggest, then, that the NH POAM–HALOE differences near 20 km in the March time periods (panels c and e in Figure 9), and thus also in the overall averages in Figure 10, are primarily due to the fact that, unlike the HALOE retrieval algorithm, the POAM retrievals do not correct for diurnal variations along the line of sight.

[24] Even when comparing the SH and NH summer periods, however, (e.g., panels a, b, and d compared to panels g and h in Figure 9) there appears to be a systematic difference between the comparisons in the two hemispheres near 20 km, where the NH POAM profiles are higher relative to HALOE than the SH profiles. *Randall et al.* [2001a] suggested that the comparisons between POAM III and SAGE II aerosol extinction profiles were consistent with a small timing error in the POAM instrument, which would correspond to a high (low)-altitude registration error of less than 175 m in the NH (SH). Simulations of such an error, however, show that at 20 km it would produce a small low bias in the POAM NH NO$_2$, and a small high bias in the POAM SH NO$_2$, the opposite of what could be concluded from the POAM–HALOE comparisons. Thus, we believe that the POAM–HALOE differences near 20 km during the summer months cannot be positively attributed to errors in either instrument individually, and most likely arise because of large uncertainties in the retrievals from both instruments, since 20 km is at the low end of the NO$_2$ retrieval altitude range.

4.2. MkIV Comparisons

[25] The MkIV interferometer is a balloon-based solar occultation FTIR spectrometer, similar to the ATMOS instrument. Thus, the accuracy of its NO$_2$ retrievals should be similar to those of the ATMOS measurements. A comparison of MkIV NO$_x$ (predominantly NO$_2$ around 20 km altitude) with in situ measurements made by instruments on board the NASA ER-2 aircraft show good (better than 10%) agreement [*Toon et al.*, 1999]. *Sen et al.* [1998] showed one comparison between the MkIV data and version 18 HALOE NO$_2$ in September 1993 that agreed to

within about 0.1 ppbv above 25 km, but only to within about 0.3–0.7 ppbv below 25 km. We have looked at the same comparison with version 19 HALOE data, and the relatively large discrepancies below 25 km are no longer present.

[26] During the SOLVE/THESEO-2000 campaign, there were two flights of the MkIV payload from the Esrange balloon facility just outside of Kiruna, Sweden, the location of which coincided closely with POAM measurements. These flights occurred on 3 December 1999, and on 15 March 2000, at the latitudes and longitudes given in Table 3. *Lumpe et al.* [2002a] showed that the MkIV ozone measured during these flights compared extremely well with coincident POAM ozone data that were acquired at similar equivalent latitudes. We describe the comparison of the NO$_2$ measurements here. As can be seen from Table 3, MkIV measured at local sunset on 3 December 1999, but at local sunrise on 15 March 2000. Since NO$_2$ exhibits significant diurnal variation in the stratosphere [e.g., *Randall et al.*, 1998, and references therein] and POAM NH measurements are all made at local sunset, only the 3 December measurements are expected to match quantitatively, so we only show these comparisons here. We note, however, that although not shown, both the POAM and MkIV profiles on 15 March have similar shapes, with generally increasing NO$_2$ mixing ratios from 20 to 30 km, and a small inflection near 25 km. MkIV densities are roughly 2 times lower than POAM, as expected from the known diurnal variations.

[27] Figure 11a shows the MkIV and POAM coincident profiles on 3 December 1999. Throughout the common altitude range, the POAM and MkIV profile shapes are quite similar. All three profiles peak near 2–3 ppbv at 31 km, dropping off to about 1 ppbv at 34 km, and to about 0 at 20 km. Nevertheless, near 31 km the POAM mixing ratios are significantly higher than the MkIV mixing ratios, and this is not expected based on the HALOE comparisons. It is unlikely that these differences are caused primarily by vortex sampling issues, as the equivalent latitude of the MkIV position was between that of the two POAM measurements from 30 to 34 km [*Lumpe et al.*, 2002a] (so variations due to vortex sampling differences would most likely have resulted in the MkIV profile lying between the two POAM profiles). Instead, we believe that the differences are caused by a geographic latitude bias in the comparisons, as discussed next.

[28] Table 3 lists the average latitude, longitude, and local time of the MkIV measurements. In fact, however, the MkIV profile was acquired over a period of about 2 hours, drifting from 67.6° to 62.7° in latitude, from 22.4° to 16.0° in longitude, and from 12.6 to 14.4 hours in local time, as the tangent altitude moved from 34 to 6 km. The drift in local time simply coincides with the drift in the local sunset
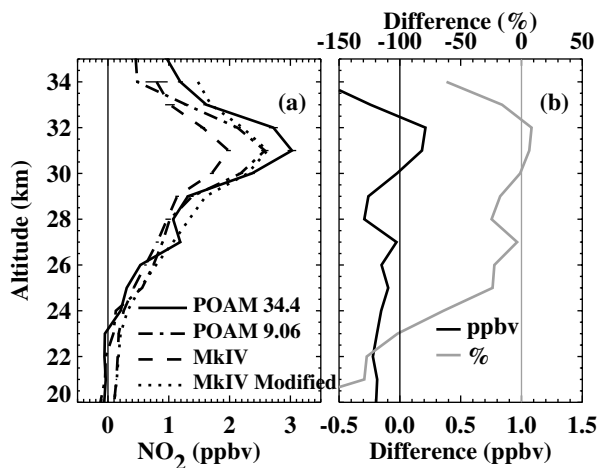
Figure 11. (a) POAM NO₂ profiles measured at 34.4°E (solid) and 9.06°E (dot-dash) compared to the MkIV profile measured at 19°E (dashed) on 3 December 1999. Error bars correspond to the quoted random errors for each instrument. The MkIV profile modified to account for the NO₂ latitudinal gradient (see text) is plotted as the dotted line. (b) The average absolute (black, bottom scale) and relative (gray, top scale) difference (equation (1)) between the two POAM profiles and the modified MkIV profile.

time as the measurement location drifted, so there should not be any true diurnal effects in the POAM−MkIV comparisons. The MkIV measurements were bracketed in longitude (and, as noted earlier, equivalent latitude) by the POAM measurements, so this also should not have been a factor in the differences. The MkIV latitude drifted such that it was poleward, by 2−3°, of the POAM measurement location for tangent altitudes near 30−33 km, but matched the POAM measurement latitude when the tangent altitude was near 20 km. This is in the correct direction for POAM to show higher mixing ratios than MkIV (i.e., NO₂ mixing ratios decrease toward higher northern latitudes in December). We have run the Naval Research Laboratory (NRL) CHEM2D model [*Siskind et al.*, 1997] to explore the magnitude of the NO₂ latitudinal gradient in December near the POAM measurement latitudes. The model predicts an exceptionally steep latitude gradient near 65°N at 30 km, with NO₂ decreases of about 0.3 ppbv for every degree increase in latitude. This strong NO₂ gradient arises from the rapid decrease of solar UV flux with latitude at the edge of the polar night, in the presence of large concentrations of N₂O₅ (>2 ppbv at 30 km, as measured by the MkIV instrument).

[29] To account for the NO₂ latitudinal gradient in the POAM−MkIV comparisons, we have modified the MkIV profile by adding to it the difference between the predicted NO₂ at the POAM latitude and that at the MkIV latitude, at each altitude, based on the NRL model. This modified profile is also shown in Figure 11a, and matches the POAM measurements much more closely. Figure 11b shows the average differences (equation (1)) between the modified MkIV profile and the two POAM profiles. The average differences are very small below 33 km, on average less than 0.2 ppbv. The two POAM profiles themselves show

differences on the order of 0.2−0.4 ppbv above 29 km. These differences correlate with differences observed in ozone [*Lumpe et al.*, 2002a], and with longitudinal variations in potential vorticity, so we attribute them to real geophysical variations. Although small, they represent a significant fraction of the difference between either of the POAM profiles and the modified MkIV profile. Note that because of the very low mixing ratios below 24 km, relative differences at these altitudes are large, and can thus be misleading regarding the qualitative agreement (since negative POAM data can cause the denominator in equation (1) to approach 0, for the calculation of relative differences we have replaced *Avg* by the MkIV data whenever the POAM data are negative).

[30] To summarize the MkIV comparisons, the differences with respect to POAM are generally about ±0.2 ppbv if one accounts for differences in the geographic location of the measurements, and some of the residual differences may also be explained by geophysical variations. Clearly, interpreting comparisons during the winter, when mixing ratios are low, and steep latitudinal and longitudinal gradients in NO₂ are present, is complex. It would be beneficial to have more comparisons between POAM and MkIV at times and locations less affected by geophysical variations. Nevertheless, based on the above analysis, we conclude that the agreement between the two instruments is quite reasonable both qualitatively and quantitatively.

4.3. LPMA/DOAS Comparisons

[31] The LPMA/DOAS instrument is a balloon-based, solar occultation, UV-visible spectrometer that flies on the LPMA/DOAS gondola. Profiles are measured as the balloon ascends through the atmosphere ("ascent" mode) as well as at float altitude (~30 km) when the Sun sets through the atmosphere ("occultation" mode). Note that both the ascent and occultation mode profiles are made via the solar occultation technique, the distinction being whether the apparent sunrise/sunset is caused by movement of the balloon or by the Earth's rotation. NO₂ is retrieved from high-resolution spectral measurements near 448 nm, very near the wavelength bands of the POAM NO₂ channels. NO₂ retrievals have an accuracy of about 2%, and an altitude resolution as high as 100 m in ascent mode and 1 km in solar occultation mode. *Boesch et al.* [1999] found that DOAS UV and visible NO₂ measurements agreed to better than ±10% with simultaneous IR solar occultation NO₂ measurements, and with measurements from the Improved Limb Atmospheric Spectrometer (ILAS).

[32] As described by *Lumpe et al.* [2002a], during the SOLVE/THESEO-2000 campaign, POAM made one measurement coincident with a flight of the LPMA/DOAS instrument from Kiruna, Sweden, on 18 February 2000 (see Table 4). The DOAS occultation NO₂ profile is compared to the coincident POAM profile in Figure 12a; the profiles are presented in concentration units since this is the native unit for both instruments. Both the DOAS and POAM profiles have similar overall shape, with maximum NO₂ densities near 24 km. At many altitudes, the predicted random error bars for the two measurements overlap. The DOAS ascent profile, which is not shown because it was measured 1.4 hours before local sunset, was also similar in shape to the POAM and DOAS occultation profiles. Ascent densities were about 20−

Table 4. DOAS/POAM Coincidences on 18 February 2000[a]

Profile	Latitude, deg	Longitude, deg east	Local time, hours
POAM	67.4	14.3	16.2
DOAS Occ	68.0[a]	21.0[a]	16.2[a]
DOAS Asc	68.0[a]	22.9[a]	14.8[a]

[a]DOAS latitude, longitude, and time correspond to the average values during the DOAS ascent or occultation.

30% lower than the DOAS occultation and POAM mixing ratios, as expected from diurnal variations in NO₂ near sunset. The POAM profile was acquired at the same local time as the DOAS occultation profile—correspondingly, the magnitude of the POAM profile more closely matches the occultation profile than the ascent profile, and this is the more appropriate comparison for quantitative validation.

[33] To quantify the POAM–DOAS differences, which are shown in Figure 12b, the DOAS occultation profile was linearly interpolated to the POAM altitude grid. To remain consistent with the HALOE and MkIV comparisons presented above, we have converted the POAM–DOAS absolute differences from density to approximate mixing ratio, by dividing the coincident DOAS and POAM data by the UKMO total atmospheric density interpolated to the POAM measurement location. At all altitudes the differences are less than ~0.2 ppbv, except at 23 km where the difference increases to 0.35 ppbv (POAM less than DOAS). This corresponds to relative differences less than 13% above 23 km, but to larger relative differences between 20 and 23 km. The large relative differences at 20 and 21 km are not a major concern, as NO₂ densities here are less than 3.0×10^8 cm⁻³ (mixing ratios less than 0.3 ppbv), and the observed differences are close to the respective uncertainties in the two measurements. We believe that the differences near 23 km might be due to dynamical factors. As noted by *Lumpe et al.* [2002a], these measurements occurred near the vortex edge, and at 23 km the equivalent latitudes were within a fraction of a degree. However, the POAM and DOAS measurements straddled the Scandinavian mountains, so that the LPMA/DOAS probed air masses in the very lee of the mountain range, whereas POAM probed air masses mostly westward (upwind) from the mountains. Therefore, differences in the measurements due to variations in the sampled air masses are to be expected, and might account for the observed profile differences near 23 km. In any case, however, the agreement shown in Figure 12 is excellent, and in accordance with the statistical results from the POAM–HALOE comparisons.

4.4. SAOZ Comparisons

[34] The SAOZ instrument is a lightweight UV-visible diode array spectrometer that measures the absorption of sunlight by the atmosphere in the ascent and occultation modes (as defined above for the LPMA/DOAS instrument). A simple conical mirror replaces the gondola orientation or Sun tracker systems generally used on large balloon platforms. The balloon version of the SAOZ instrument is very similar to the one used for ground-based measurements of total ozone and NO₂ [*Pommereau and Goutail*, 1988]. Measurements are made between 290 and 640 nm, with an average resolution of 0.8 nm. Like the DOAS instrument, NO₂ is measured in the spectral region near 448 nm (from 410 to 530 nm) using cross sections measured at 220 K by

Vandaele et al. [1998] (3% accuracy). The spectral analysis and the inversion scheme used for SAOZ are discussed by *Pommereau and Piquard* [1994]. The average precision in winter Arctic conditions derived from the spectral fit varies from 0.4×10^8 mol cm⁻³ (8%) at 24 km, to 0.6×10^8 mol cm⁻³ (20%) at 21 km, to 0.78×10^8 mol cm⁻³ (37%) at 17 km. Use of constant-temperature (220 K) cross-sections results in a high bias of 4%, 6%, and 8% at 24, 21, and 17 km, respectively, for an average temperature gradient (L. Denis, J.-P. Pommereau, F. Goutail, and F. Lefevre, Denitrification, NOₓ redistribution and chlorine activation in the winter Arctic vortex of 1997–2000 from SAOZ UV-visible measurements on short and long duration balloons, in preparation). The SAOZ is equipped with a Global Positioning System (GPS) receiver, which allows its location to be determined in three dimensions with an uncertainty of ±150 m. The vertical resolution of the SAOZ NO₂ profiles is 1 km. The SAOZ NO₂ data shown below have not been corrected for diurnal variations along the line of sight, consistent with the POAM data.

[35] *Lumpe et al.* [2002a] described comparisons between SAOZ and POAM III O₃ profile measurements from the SOLVE/THESEO 2000 campaign. They showed that the differences between the two instruments were on average within 5–7% between 14 and 27 km. Here we show comparisons of POAM NO₂ profiles to those measured by the SAOZ instrument during the same SOLVE/THESEO 2000 flights described by *Lumpe et al.* [2002a], as well as during a few earlier flights. Table 5 lists the relevant information for the nine flights during which the SAOZ measurements were coincident with POAM measurements. One flight (17 November 1999) originated in Andoya, Norway (69°N, 16°E), and the other eight flights originated in Kiruna, Sweden (68°N, 21°E). Like for the DOAS comparison, we only include comparisons with SAOZ occultation data, which all occurred at local sunset and were thus coincident in time with the POAM measurements. Table 5 lists the midpoint latitude, longitude, and time for
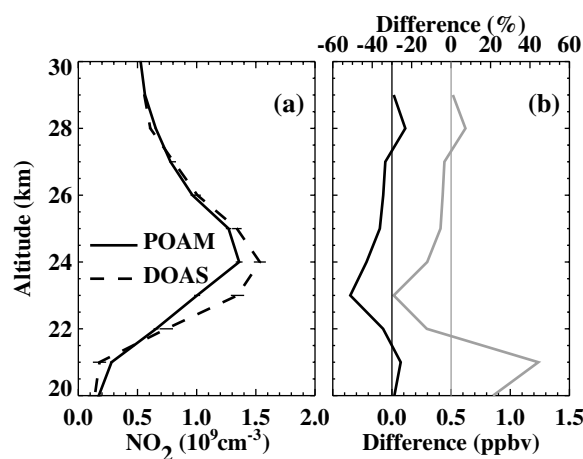

Figure 12. (a) Coincident NO₂ profiles measured by POAM (solid) and the LPMA/DOAS instrument (dashed) on 18 February 2000. Error bars correspond to the quoted random errors for each instrument. (b) Absolute (black, bottom scale) and relative (gray, top scale) POAM–DOAS differences (equation (1)).

Table 5. SAOZ/POAM Coincidences[a]

Date	SAOZ Latitude, deg	POAM Latitude, deg	SAOZ Longitude, deg	POAM Longitude, deg	SAOZ Time, hours	POAM Time, hours
27 January 1999	65.00	65.81	17.20	9.990	14.00	14.33
6 February 1999	66.20	66.75	17.60	16.04	14.40	14.44
18 February 1999	66.60	67.55	16.20	9.500	15.30	15.59
17 November 1999	67.20	66.72	14.40	11.93	13.10	13.41
28 January 2000	65.60	65.84	18.80	20.52	13.90	13.67
13 February 2000	67.20	67.21	18.90	10.61	14.70	15.21
27 February 2000	67.00	67.75	18.60	16.19	15.60	15.70
7 March 2000	66.80	67.68	15.20	18.13	16.40	16.14
25 March 2000	68.70	66.53	15.00	20.85	17.50	17.02

[a]Columns: (1) coincidence date, (2) SAOZ latitude at 20 km, (3) POAM latitude, (4) SAOZ longitude at 20 km, (5) POAM longitude, (6) SAOZ local time at 20 km, and (7) POAM local time.

each coincident SAOZ occultation measurement. On average the POAM data were acquired 0.2° North and 2.5° West of the SAOZ occultation data.

[36]   The average POAM and SAOZ NO$_2$ profiles for the group of coincidences listed in Table 5 are shown in Figure 13a. The shapes of the profiles measured by both the POAM and SAOZ instruments are similar, with gradually increasing concentrations from 20 to 23 km, and nearly constant concentrations above this. The statistical differences between the POAM and SAOZ data are shown in Figure 13b. Again, to remain consistent with the other comparisons presented here, we have converted the POAM−SAOZ density differences to approximate mixing ratio differences by dividing the POAM and SAOZ NO$_2$ densities by the UKMO total atmospheric density interpolated to the coincident POAM measurement locations. The two instruments agree quite well, with average differences less than 0.25 ppbv at all altitudes except 27 km, where the average difference of 0.49 ppbv has a fairly large statistical uncertainty of 0.33 ppbv. The relative differences shown in Figure 13 are only marginally significant above 22 km, but approach 70% at 20 km; this simply reflects the very low wintertime mixing ratios characteristic of these comparisons (note that there were three negative POAM measurements at 20 km, and one at 21 km, for which we have replaced *Avg* by the SAOZ data in equation (1)).

[37]   All of the POAM−SAOZ comparisons occurred during the NH vortex season, when geographic separations can significantly affect the agreement among different measurements. As pointed out by *Lumpe et al.* [2002a], all of the SOLVE/THESEO 2000 comparisons except one (27 February) occurred where both instruments were sampling similar equivalent latitudes. The measurements that occurred on 27 February 2000 and those during winter 1999 were all placed near the edge of the vortex, where the SAOZ and POAM instruments were more likely to sample air parcels with different histories. The character of the average difference profile presented here is not significantly changed, however, when these vortex edge comparisons are omitted from the analysis.

4.5.   NIWA Column Comparisons

[38]   The correlative measurements described above offer the potential for comparing NO$_2$ profiles. In this section we compare vertically integrated POAM NO$_2$ data to the total NO$_2$ column measured from the ground. The column NO$_2$ measurements used for this purpose were made by NIWA at Arrival Heights, Antarctica (77.8°S, 166.6°E), and at Kiruna,

Sweden (67.8°N, 20.4°E). The Swedish Institute of Space Physics operates the NIWA UV/visible spectrometer at Kiruna. The NO$_2$ columns are derived from differential absorption spectroscopy of sunlight scattered from the zenith sky at dawn and dusk. The analysis method used is identical for both instruments and is similar to that described by *Hofmann et al.* [1995] and *Harder et al.* [1997]. Since the measurements are made using the zenith sky viewing technique, the measured NO$_2$ slant columns must be converted into vertical columns for comparison with the POAM data. This is accomplished with a radiative transfer model based on the work of *Frank and Platt* [1990]. The measurements are made at both sr and ss at solar zenith angles from 80° to 95°, in steps of 1°. For all comparisons with POAM we use NIWA data obtained at a solar zenith angle of 90°, except at Kiruna from 3 December to 8 January, for which we use data obtained at a solar zenith angle of 92° (90° measurements were not possible because of continual darkness at the surface at this time).

[39]   The Network for the Detection of Stratospheric Change (NDSC) has conducted two intercomparisons of different NO$_2$ column measurements, in 1992 [*Hofmann et al.*, 1995] and in 1996 [*Roscoe et al.*, 1999]. In the 1996
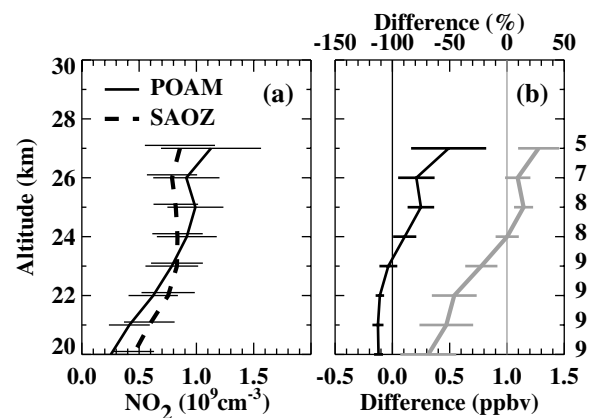

Figure 13.   (a) Average POAM (solid) and SAOZ (dashed) NO$_2$ profiles for all coincidences (see Table 5). "Error" bars represent the 1σ uncertainty in the mean profiles and are offset high by 0.1 km on the SAOZ profile for clarity. (b) Average absolute (black, bottom scale) and relative (gray, top scale) differences (equation (1)) for the coincidences in Table 5. "Error" bars represent the 1σ uncertainties in the mean differences. The numbers of coincident data points are given on the right vertical axis.
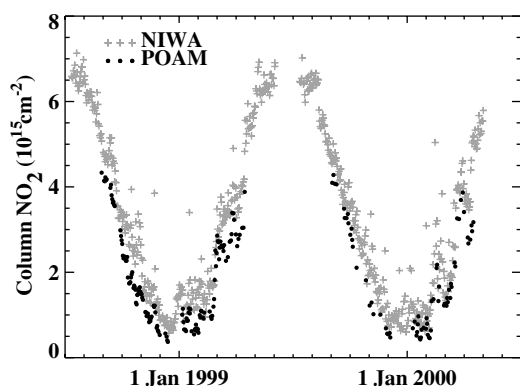
Figure 14. Column NO_2 measured by the NIWA UV spectrometer (gray plus marks) and POAM (black dots, 20–45 km) above Kiruna, Sweden. All measurements were made at local sunset.

intercomparison, there were differences greater than 25–30% between the extremes of the 15 instruments for the total column NO_2 on most of the days. When averaged over the entire intercomparison period, the mean difference between most of the instruments was ±6% (1σ). The NIWA instrument was generally near the middle of this range. *Roscoe et al.* [1999] developed a regression analysis method to compare the different instruments when no one instrument was known to be better than any of the others. They noted that the NIWA instrument was one of three instruments to show "consistently smaller residual errors" in this regression analysis.

[40] Figure 14 compares the temporal variation in column NO_2 measured by the NIWA instrument located at Kiruna, Sweden to the partial column NO_2 calculated from POAM data between 20 and 45 km. Only those POAM measurements that were within 4° in latitude and 12° in longitude of Kiruna are shown. Since all of the POAM data were acquired at local sunset, we only show NIWA data acquired at sunset. The POAM data track the relative temporal variations in NO_2 as observed by the NIWA instrument extremely well, including even many short-term variations (e.g., December 1998–February 1999, March–April 1999, and February–March 2000).

[41] Figure 14 clearly shows that the POAM values are systematically lower than the NIWA data. A low bias is expected in the POAM data since the column was calculated only from 20 to 45 km, and thus does not include the tropospheric, lower stratospheric or upper stratospheric NO_2 seen by the NIWA instrument. There are also a number of NIWA data points in the winter seasons that fall well above the average column values defined by the majority of the data, when the POAM data show no such excursions. We ascribe these high values to high tropospheric pollution that would not be seen in the POAM data; this is consistent with the works of *Elokhov and Gruzdev* [1995], who observed a preponderance of such events during the winter over the Zvenigorod station (56°N) near Moscow, and of *Pommereau and Goutail* [1988], who observed wintertime pollution episodes over Sodankyla, Finland (67°N).

[42] To quantify the low bias due to the lack of tropospheric and lower stratospheric contribution to the POAM

data, we have calculated the differences between the POAM and NIWA measurements in Figure 14 that occurred within 2 hours of each other. Figure 15 shows these differences as averages within bins defined by the NIWA column NO_2, each bin centered on 5×10^{14} cm^{-2} increments, with a width of $±2.5 \times 10^{14}$ cm^{-2}. The coincident data set used for Figure 15 included a total of 157 measurements, with 10–35 measurements in each bin. The average differences (POAM–NIWA) in time, latitude and longitude were $+0.5 ± 0.4$ hours, $-0.5° ± 2.0°$, and $-0.2° ± 7°$, respectively. We examined the data for possible correlations between the observed POAM–NIWA column NO_2 differences and the latitude, longitude or temporal separation of the coincident event pairs, but did not find robust correlations with any of these parameters. The overall average relative (absolute) column NO_2 difference is $-37.4 ± 1.7\%$ $(-7.5 ± 0.3 \times 10^{14}$ cm$^{-2})$, where the negative difference signifies that POAM was lower than NIWA, and the errors refer to the 1σ uncertainty in the means.

[43] The standard climatological NO_2 profile from the work of *Anderson et al.* [1986] yields a column total from 0 to 50 km of 5.1×10^{15} mol cm^{-2}, of which 1.3×10^{15} mol cm^{-2} (25%) lies below 20 km, and about 1% above 45 km. Based on these numbers, we should expect the POAM data to underestimate the NIWA observations by about $1.3–1.4 \times 10^{15}$ mol cm^{-2}, or ~26%. As shown in Figure 15 the differences between POAM and the NIWA measurements vary from about 0.3 to 1.0×10^{15} mol cm^{-2}, or about 15–45%. None of these comparisons were made, however, at a time/region where the total column from the NIWA measurements was as high as the climatological value from the work of *Anderson et al.* [1986]. The absolute (relative) differences in Figure 15 generally decrease (increase) with increasing column NO_2. A linear extrapolation of the differences to a column NO_2 value of 5.1×10^{15} mol cm^{-2} yields a rough approximation of the differences expected if the column NO_2 were equal to the climatological value. The absolute (relative) extrapolated value is $1.0 ± 0.3 \times 10^{15}$
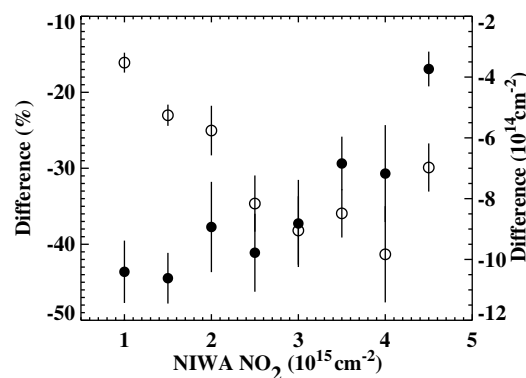

Figure 15. Average column NO_2 differences (equation (1)) between the POAM and NIWA coincidences over Kiruna, Sweden, shown as % (black dots, left scale) and in absolute units (POAM–NIWA, open circles, right scale). The averages are calculated for all coincidences where the NIWA column NO_2 lay within $±2.5 \times 10^{14}$ cm^{-2} of the values plotted. "Error" bars denote the 1σ uncertainty in the mean differences in each bin.
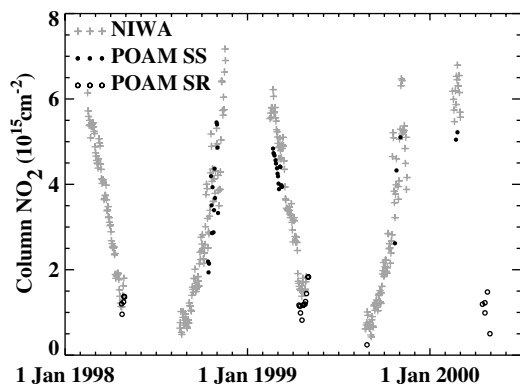
Figure 16. Column NO$_2$ measured by the NIWA UV spectrometer (+) and POAM (20–45 km) at local sunrise (open circles) and at local sunset (dots) above Arrival Heights, Antarctica. Only NIWA measurements made at local sunrise are plotted from 9 April to 8 September, and only local sunset measurements are plotted at other times, to correspond to the local times of the POAM data.

mol cm^{-2} (19 ± 7%), close to the expectation from the climatological reference.

[44] Figure 16 is analogous to Figure 14, but shows the column NO$_2$ measurements made by the POAM and NIWA instruments over Arrival Heights, Antarctica. In the SH, POAM makes measurements at local sunrise from early April to early September, and at local sunset at all other times—thus, the figure only includes the corresponding sunrise/sunset NIWA data. There are many fewer comparisons at this station, but the results are consistent with the Kiruna results, showing qualitatively similar column variations from both instruments. There were a total of 36 coincident measurements, including 28 made at local sunset and 8 made at local sunrise. For the sunset coincidences the average time, latitude and longitude separations (POAM–NIWA) were −0.5 ± 0.8 hours, −0.3 ± 2°, and −0.2 ± 7°, respectively. The analogous quantities for the sunrise coincidences were +0.2 ± 0.8 hours, −0.9 ± 2°, and −1 ± 8°, respectively. As for the Kiruna comparisons, we found no robust systematic variations in the differences based on the spatial or temporal separations. The average relative (absolute) NO$_2$ column difference was −23.2 ± 2.6% (−7.1 ± 0.8 × 10^{14} cm^{-2}) for the combined sunrise and sunset coincidence data sets, close to the value expected based on the *Anderson et al.* [1986] climatology. When examined separately, the average difference for the local sunrise events, which all occurred during winter when stratospheric NO$_2$ was relatively low, was −37 ± 6% (−4.8 ± 0.7 × 10^{14} cm^{-2}). The average relative (absolute) difference for the sunset coincidences, which occurred during spring and fall when column NO$_2$ was relatively high and closer to the climatological value, was −19 ± 3% (−7.7 ± 0.9 × 10^{14} cm^{-2}).

[45] The comparisons shown in Figures 14, 15, and 16 show that the partial column data calculated from POAM NO$_2$ agree with the NIWA column measurements in terms of the temporal variation, and are quantitatively within expectations. This was expected based on the good agreement in the profile comparisons. Work is in progress to determine if comparisons such as these can be used to infer

information regarding the variations in tropospheric and lower stratospheric NO$_2$.

5. Summary

[46] POAM III NO$_2$ densities are retrieved from 20 to 45 km, with a vertical resolution of about 1.5–2.5 km at altitudes below 40 km, degrading to more than 7 km at an altitude of 45 km. The comparisons described above show generally very good agreement between POAM and the correlative measurements. Although there were too few coincidences to constitute statistically significant comparisons for many of the correlative data sets, we have demonstrated that POAM measurements of NO$_2$ are reasonable in terms of their magnitude, profile structure, and temporal variations. Figure 17 summarizes the differences between POAM and the correlative measurements. Below about 35 km, there is no clear evidence in the DOAS, SAOZ and MkIV comparisons for a significant POAM bias. Although large relative differences are shown near 20 km, there is a large variation in their magnitude and sign, and they correspond to mixing ratio differences that are all less than 0.2 ppbv.

[47] Because of the numerous coincidences between HALOE and POAM, these satellite comparisons offer the best opportunity for detailed analysis of possible systematic errors in the POAM NO$_2$ measurements. Comparisons with nearly 200 HALOE profiles in each hemisphere show that on average POAM and HALOE agree to within about 0.2 ppbv from 20 to 33 km, or to within about 6% at most of these altitudes. Relative differences are larger near 20 km where the mixing ratios become very small. These differences result mainly from the fact that the POAM retrievals do not correct for diurnal variations along the line of sight, whereas this correction is an integral part of the HALOE retrievals. POAM is consistently higher than HALOE between about 35 and 42 km, by up to about 0.7 ppbv (17%). We have identified a systematic error in the HALOE retrievals resulting from improper separation of the aerosol and NO$_2$ contributions in the NO$_2$ channel that accounts for about 30% of this POAM–HALOE difference, leaving a maximum residual difference of about 12% near 40 km that
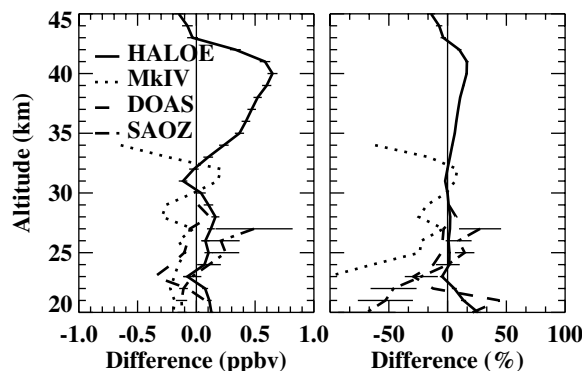

Figure 17. Average absolute (left) and relative (right) differences (equation (1)) between POAM and HALOE (solid), MkIV (dotted), DOAS (dashed), and SAOZ (dot-dash) for coincidences listed in Tables 2, 3, 4, and 5. "Error" bars represent the 1σ uncertainties in the mean differences for comparisons that included multiple profiles.

is not currently understood. We emphasize that this difference is relatively small considering the state of remote NO$_2$ measurements today. Differences decrease above 40 km, and are slightly negative (0.1–0.2 ppbv on average) at 45 km. This is most likely due to an increasing dependence of the POAM retrievals on the a priori NO$_2$ profile, and the decreasing altitude resolution of the POAM measurements. We have shown that some sunspots and very high aerosol extinction, as might be found in the presence of PSCs, can cause artifacts in the NO$_2$ retrievals. We recommend that the POAM NO$_2$ data be screened for these effects, as described above, before the data are analyzed for scientific purposes. With the above considerations, we conclude that the POAM III NO$_2$ profiles from 20 to 45 km constitute a robust data product that is appropriate for scientific analysis and for the validation of NO$_2$ measurements from other instruments.

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