Global, Long-Term Sulphur Dioxide Measurements From TOVS Data: A New Tool for Studying Explosive Volcanism and Climate

A. J. Prata1, W. I. Rose2, S. Self3, and D. M. O’Brien1

A new technique for retrieving sulphur dioxide concentrations from TIROS Operational Vertical Sounder (TOVS) data is described. The retrieval technique relies on absorption of infrared radiation by the anti-symmetric stretch of the sulphur dioxide (SO2) molecule centred around 7.3 µm. The High-resolution infrared radiation sounder (HIRS/2) is part of the TOVS package and has a channel that covers this absorption region. The HIRS/2 data are global, span almost 24 years, have a sub-satellite spatial resolution of about 18 km and can be used both day and night. The retrieval method is described and its accuracy and sources of error discussed. Case studies for the June 1991 Pinatubo eruptions, for the August 1991 eruptions of Cerro Hudson and for several eruptions of Hekla volcano, are used to illustrate the retrievals and the results are compared with independent SO2 retrievals from the TOMS instrument. These new SO2 data provide a potentially valuable tool for studying the climatic effects of explosive eruptions. Because the satellite measurements are global, long-term and can simultaneously provide other climate parameters (e.g. surface temperatures, temperature profiles, humidity profiles, cloudiness, ozone amount and long- and short-wave radiation) they can be used to test and validate volcanically-induced effects in global climate simulations.

1. INTRODUCTION

The global atmospheric budget of sulphur is important to climate [Seinfeld and Pandis, 1998]. Knowledge of the SO2 component in the atmosphere has come from a variety of sources including ground-based, airborne and more recently satellite-based measurements [Stoiber and Jepsen, 1973; Allard et al., 1994; Pyle et al., 1996; Bluth et al., 1993; Bluth et al., 1997]. Estimates of the SO2 budget provide limits on the amount of H2SO4 aerosol in the stratosphere [Hofmann and Rosen, 1981], which in turn can be used to evaluate the radiative effects on climate [Kiehl and Briegleb, 1993; Mass and Portman, 1989; Robock, 2000]. Stratospheric sulfuric acid aerosol derives principally from SO2 injected by volcanic eruptions and these aerosols are important in the chemistry of the stratosphere by providing surfaces for chemical reactions. The current estimate of the stratospheric portion of the flux of SO2 ranges from ~0.3 Tg a−1 to ~3 Tg a−1, with a mean of ~1 Tg a−1 [Pyle et al., 1996].

Indirect measurements of this flux have been made from the Total Ozone Mapping Spectrometer (TOMS) satellite [Bluth et al., 1997; Krueger et al., 1995], and more recently from the Global Ozone Monitoring Experiment (GOME) — see for example Eisinger and Burrows [1998]. Rose et al. [2000] have shown the usefulness of combining different satellite measurements of SO2 to improve estimates of the atmospheric emission of this gas from volcanic eruptions. Given the importance of the stratospheric budget of SO2 to climate and the sporadic and global nature of vol-
canic eruptions, it is useful to have other independent satellite measurements of SO\textsubscript{2}. The purpose of our work here is to document a source of satellite SO\textsubscript{2} measurements that complement existing measurements and provide a valuable data-set for intercomparison with TOMS and GOME.

The NOAA series of satellites carry the TIROS Operational Vertical Sounder (TOVS) instrument package which includes the High-resolution Infrared Radiation Sounder (HIRS/2—see Smith et al., [1979]). This instrument measures infrared radiation in nineteen narrow channels (bandwidths =0.5–1 \(\mu\)m) with the purpose of retrieving atmospheric temperature and moisture profiles. One channel (channel 11) completely covers the strong, anti-symmetric stretch vibration (\(v_3\)-band) of SO\textsubscript{2} situated near 7.34 \(\mu\)m (see Figure 1). This channel is used for tropospheric water vapour sounding, but it is shown here that provided the water vapour lies below the SO\textsubscript{2}, the channel can also be used to measure SO\textsubscript{2} column abundance.

Figure 1 shows the vertical transmittance, in the wavelength range 6–13 \(\mu\)m, for a cloud layer containing 100 m atm-cm (dashed line) and 1000 m atm-cm (solid line) of SO\textsubscript{2}, with a width of 2 km, placed at 25 km in an otherwise absorber-free atmosphere.

The main absorption features are the \(v_1\) and \(v_3\) bands, centred near 8.55 \(\mu\)m and 7.34 \(\mu\)m, respectively. Both of these bands can be used to retrieve SO\textsubscript{2} column amounts.\(^1\) The \(v_1\) feature looks particularly interesting because it lies within the ‘window’ where absorption due to water vapour is smaller. Indeed Realmuto et al., [1994] and Realmuto [1996] have developed methods for retrieving SO\textsubscript{2} using this feature from airborne and spaceborne sensors (e.g. MASTER, ASTER and MODIS). Ground-based interferometric measurements have also been made using this band [Francis et al., 1995; Love et al., 1998; McGee and Gerlach, 1998]. Use of the \(v_3\)-band can be traced to work first published by Mankin et al. [1992] and Goldman et al. [1992] who used a high spectral resolution instrument (an interferometer) to exploit the ‘micro-windows’ around 7.3 \(\mu\)m. Crisp [1995] was first to suggest that the 7.3 \(\mu\)m band on MODIS could be used to provide an SO\textsubscript{2} alert, but she did not develop a quantitative retrieval procedure. Ackerman and Strabala [1994] used HIRS/2 data to study \(\text{H}_2\text{SO}_4\) aerosols by utilising information in the 8- to 12-\(\mu\)m region. Baran and Foot [1994] demonstrated the use of HIRS/2 data and radiative transfer modelling to derive quantitative estimates of \(\text{H}_2\text{SO}_4\) aerosols. Our work is the first to attempt a quantitative retrieval based on broadband (bandwidths \(>\sim 0.4\) \(\mu\)m) infrared measurements of the \(v_3\) absorption feature of SO\textsubscript{2}. In essence the retrieval from satellite-based broadband measurements is possible whenever the SO\textsubscript{2} is well-separated from H\textsubscript{2}O and above it. Thus the method is ideally suited to studies of SO\textsubscript{2} that affect the climate, that is, quantitative studies of large volcanic emissions of SO\textsubscript{2} that have been injected into the upper troposphere/lower stratosphere.

In this paper we describe the principle of the retrieval method and suggest a model and methodology for use with HIRS/2 data. Next we discuss the practical implementation of the algorithm and provide a thorough error analysis. Examples of retrievals for several well-studied eruptions are given together with results for the 1996 Mt Ruapehu eruptions—for which no TOMS data were available. Two appendices are provided—Appendix 1 contains a list of the acronyms used throughout the paper, and Appendix 2 contains some of the mathematical detail. Finally, we suggest ways these new data can be used to study the atmospheric effects of explosive volcanism on climate.

2. SO\textsubscript{2} RETRIEVAL FROM INFRARED MEASUREMENTS

2.1 The HIRS/2 Instrument

The first TOVS package was launched on board the TIROS-N satellite in 1978 and the latest is on the NOAA-17 platform—providing 24 years of continuous global measurements. The HIRS/2 instrument includes measurements in 20 channels (19 in the infrared and 1 visible channel)—giving estimates of surface temperature, atmospheric profiles of temperature and moisture, cloudiness and column ozone abundance. These data are used operationally to provide meteorological data for numerical weather forecasting.
The channels were chosen to maximize information retrieved from radiances sampling different vertical layers of the atmosphere several kilometres thick. Most of the channels lie near strong absorption features (e.g. near to CO\textsubscript{2} or H\textsubscript{2}O absorption features), while some channels are located in atmospheric window regions where information about the surface and lower atmosphere can be retrieved. Table 1 provides information about the channels of relevance to this study. Table 2 provides information on the HIRS/2 instrument—more comprehensive information may be found in Smith et al. [1979], Planet [1988], and Kidwell [1991].

The filter functions of the HIRS/2 instrument are also shown in Figure 1, together with SO\textsubscript{2} transmittance of the \textit{v}\textsubscript{1} and \textit{v}\textsubscript{3} SO\textsubscript{2} bands. Only the channels at 6.7, 7.3, 8.2, 9.7, 11 and 12 \textmu m are shown—the channels used in the retrieval are plotted using thick lines. Note that the SO\textsubscript{2} \textit{v}\textsubscript{3} feature is almost entirely covered by HIRS/2 channel 11, while only a small part of the SO\textsubscript{2} \textit{v}\textsubscript{1} band is covered by channel 10. The HIRS/2 instrument has undergone some changes over its 24 year history; there are in fact three versions of the instrument—HIRS (originally flown on Nimbus-6), HIRS/2 (NOAA-satellite series) and the present HIRS/3. Table 3 provides a chronology of the instruments’ service record, spacecraft, launch date, and the node crossing times.

### 2.2 Transmittance Model

To exploit the SO\textsubscript{2} information contained in the HIRS/2 measurements we employ an accurate transmittance model that relates the absorption in HIRS/2 channel 11 to SO\textsubscript{2} absorber amount. The anti-symmetric stretch of the SO\textsubscript{2} molecule is situated around 7.34 \textmu m and the transmittance of this feature has been modelled using a double-exponential model by Pierluissi and Tomiyama [1980] and Pierluissi et al. [1984]. We validated this model against line-by-line calculations and found the results to be accurate below about 200 m atm-cm\textsuperscript{2}. Above this value the model diverged from the line-by-line results. Based on this assessment we decided to use an exponential sum-fitting procedure (ESFT, see Wiscombe and Evans [1977]) to the band transmittance. The ESFT model has the form,

\[ t_\ell = \sum_{i=1}^{n} a_i \exp(-k_i u), \]

where \( t_\ell \) is the transmittance (see Appendix 2 for a definition), \( u \) is absorber amount, and \( a_i, k_i \) are coefficients. This model is accurate over a wide range of absorber amounts and is very fast for use operationally because a table of coefficients can be generated off-line for each assumed absorber layer (characterized by its atmospheric layer pressure and temperature) and for all the satellite filter response functions. Since HIRS/2 channel 11 has a finite bandpass, the coefficients for the band model account for the variation of the absorption across the channel 11 filter function. In practice the absorber amounts are pre-computed for 26 different atmospheric layers, and each HIRS/2 channel 11 on each satellite, in 0.001 transmittance steps. This provides a large look-up table which can be efficiently searched to provide excellent processing.

### Table 1. Characteristics of the HIRS/2 channels relevant to SO\textsubscript{2} retrieval

<table>
<thead>
<tr>
<th>Channel No.</th>
<th>Central wavelength (\textmu m)</th>
<th>Half-power bandwidth (\textmu m)</th>
<th>Purpose</th>
<th>Weighting function peak (hPa); (km in brackets)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>11.11</td>
<td>0.45</td>
<td>Surface/clouds</td>
<td>Surface (0–2 km)</td>
</tr>
<tr>
<td>9</td>
<td>9.71</td>
<td>0.48</td>
<td>Ozone</td>
<td>25 (25–35 km)</td>
</tr>
<tr>
<td>10</td>
<td>8.16/12.47\textsuperscript{a}</td>
<td>0.42/0.46\textsuperscript{b}</td>
<td>Surface/cloud</td>
<td>900 (1–2 km)</td>
</tr>
<tr>
<td>11</td>
<td>7.33</td>
<td>0.44</td>
<td>Water vapour</td>
<td>700 (3–4 km)</td>
</tr>
<tr>
<td>12</td>
<td>6.72/6.52\textsuperscript{a}</td>
<td>0.37/0.37\textsuperscript{b}</td>
<td>Water vapour</td>
<td>500 (5–6 km)</td>
</tr>
</tbody>
</table>

\textsuperscript{a} New central wavelength on HIRS/3.
\textsuperscript{b} New half-power bandwidth on HIRS/3.

### Table 2. Characteristics of the HIRS/2 instruments.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Field-of-view</td>
<td>21.8 mrad</td>
</tr>
<tr>
<td>Ground field-of-view Nadir</td>
<td>18.5 km circular</td>
</tr>
<tr>
<td>End of scan</td>
<td>31.8 km × 62.8 km</td>
</tr>
<tr>
<td>Scan line</td>
<td></td>
</tr>
<tr>
<td>Scan time</td>
<td>6.4 s</td>
</tr>
<tr>
<td>Scan angle</td>
<td>±49.5º</td>
</tr>
<tr>
<td>Swath width</td>
<td>±1115 km</td>
</tr>
<tr>
<td>Number of spots per scan line</td>
<td>56</td>
</tr>
<tr>
<td>Distance between spots Cross-track</td>
<td>26.4 km</td>
</tr>
<tr>
<td>Along-track</td>
<td>41.8 km</td>
</tr>
<tr>
<td>Gap between consecutive passes at the equator</td>
<td>540 km</td>
</tr>
</tbody>
</table>
speed for global retrievals. The precise look-up tables used are available from the authors on request.

A simplified radiative transfer model is proposed for deriving SO₂ transmittance from the HIRS/2 temperature measurements. The details of the derivation are given in Appendix 2—the main result is given here. As noted in Appendix 2, Eq. (16) may be written in the form:

\[ I - I_a = (1 - t_s) (B_s - I_a), \]

where \( I \) is the measured radiance at 7.3 \( \mu \)m, \( I_a \) is the radiance at 7.3 \( \mu \)m for an atmosphere free of SO₂, \( t_s \) is the SO₂ transmittance and \( B_s \) is the Planck radiance for the SO₂ cloud layer at temperature \( T_s \). This equation suggests that,

\[ \Delta T = \alpha + \beta (1 - t_s). \]

where \( \Delta T \) is the temperature difference corresponding to the difference between the measured radiance and the SO₂-free radiance, obtained using the inverse Planck function. \( \alpha \) and \( \beta \) are parameters to be estimated. From analysis of data and from Modtran [Berk et al., 1989] modelling it has been found that,

\[ \alpha = -8 K, \quad \beta = -32 K. \]

In practice \( \alpha \) and \( \beta \) are functions of the atmospheric state and, in particular the temperature of the SO₂ cloud. Since the SO₂ cloud temperature is unknown we have used data and modelling to estimate these parameters, but the algorithm still requires an independent estimate of the height of the SO₂ cloud in order to generate the appropriate look-up table.

### 3. PRACTICAL CONSIDERATIONS

The simple model presented here permits the evaluation of \( t_s \) from a measurement of \( \Delta T \). For HIRS/2 data, the evaluation of \( \Delta T \) is made by estimating \( I_a \) through a linear interpolation of a radiance measurement at 6.7 \( \mu \)m (HIRS/2 channel 12) and a radiance measurement at 11.1 \( \mu \)m (HIRS/2 channel 8). The use of the 11.1 \( \mu \)m channel is made in preference to a channel at 8.2 \( \mu \)m because this channel is also affected by SO₂ absorption and so cannot give a reliable estimate of the unperturbed atmospheric state. Thus,

\[ \Delta = I_{a, lin} - I_{a, lin} \]

Finally,

\[ \Delta I_{a, lin}^b = B[T, \lambda_{7.3}] - I_{a, lin}^b. \]

It should be noted that, in general, \( \Delta I_{a, lin}^b < 0 \). The HIRS/2 data processing proceeds on a pixel-by-pixel basis.

### 3.1 Estimating the Background Radiance

The scheme outlined above relies on an estimate of the background (unperturbed) radiance at 7.3 \( \mu \)m, which has been obtained using a linear interpolation (Eq. 5). The accuracy of the linear interpolation of radiance as a function of wavelength has been gauged by studying large amounts of HIRS/2 data for cases where there are no SO₂ clouds present. In most situations this appears to be accurate (within ±2

#### Table 3. Chronology of the NOAA meteorological satellite series.

<table>
<thead>
<tr>
<th>Space-craft</th>
<th>Launch date</th>
<th>Ascending node</th>
<th>Descending node</th>
<th>Service period</th>
</tr>
</thead>
<tbody>
<tr>
<td>NOAA-9</td>
<td>Dec-12-1984</td>
<td>14:20</td>
<td>02:20</td>
<td>Feb-25-1985-Aug-03-1995</td>
</tr>
<tr>
<td>NOAA-10</td>
<td>Sep-17-1986</td>
<td>19:30</td>
<td>07:30</td>
<td>Nov-17-1986-March-1995</td>
</tr>
<tr>
<td>NOAA-12</td>
<td>May-14-1991</td>
<td>19:30</td>
<td>07:30</td>
<td>May-14-1991-present</td>
</tr>
<tr>
<td>NOAA-13</td>
<td>Aug-09-1993</td>
<td></td>
<td></td>
<td>Power failure 12 days after launch</td>
</tr>
<tr>
<td>NOAA-14</td>
<td>Dec-300-1994</td>
<td>13:40</td>
<td>01:40</td>
<td>Dec-30-01994-present</td>
</tr>
<tr>
<td>NOAA-16</td>
<td>Sep-21-2000</td>
<td>14:00</td>
<td>02:00</td>
<td>Sep-21-2000-present</td>
</tr>
<tr>
<td>NOAA-17</td>
<td>Jun-24-2002</td>
<td>22:00</td>
<td>10:00</td>
<td>Jun-24-2002-present</td>
</tr>
</tbody>
</table>
K in 95% of cases studied). There is however a notable bias with the interpolated value usually higher than the actual value. In some cases, with no SO$_2$ present, it was found that the linear interpolation was a poor estimate of the observed radiance. Presumably, this is due to the effects of clouds within the HIRS/2 field-of-view. With just a small addition in complexity, a significant improvement to the scheme was made in the following manner:

1. Blackbody radiance curves are generated using the observed radiances at 6.7 and 11.1 µm, and ‘pseudo’ radiances ($I^*(T_{λ_1}, λ_2)$) are calculated at all three wavelengths:

   \[
   I' \left( T_{6.7}, 6.7 \right) = B(T_{6.7}, 6.7),
   \]

   \[
   I' \left( T_{7.3}, 7.3 \right) = B(T_{7.3}, 7.3),
   \]

   \[
   I' \left( T_{11.1}, 7.3 \right) = B(T_{11.1}, 7.3),
   \]

   \[
   I' \left( T_{6.7}, 11.1 \right) = B(T_{6.7}, 11.1),
   \]

   The blackbody radiance curves bracket the range of possible observed radiances at 7.3 µm. (See Figure 2.)

2. An estimate of the observed radiance is obtained from:

   \[
   \hat{I}_α = 0.5 \left[ I' \left( T_{6.7}, 7.3 \right) + I' \left( T_{11.1}, 7.3 \right) \right].
   \]

3. An adjustment to the linearly interpolated 7.3 µm radiance is obtained from:

   \[
   I^\text{adj}_α = 2.0 \left[ I' \left( T_{11.1}, 7.3 \right) - I' \left( T_{6.7}, 7.3 \right) \right].
   \]

4. If the brightness temperature at 11.1 µm is above a certain threshold (270 K is used), then the 7.3 µm radiance estimate is obtained from the adjusted linearly interpolated value; otherwise the average of the ‘pseudo’ radiances is used:

   \[
   \hat{I}_{7.3} = I^\text{adj}_α \quad T_{11.1} > T_{\text{thresh}},
   \]

   \[
   \hat{I}_α = 0.5 \left[ I' \left( T_{6.7}, 7.3 \right) + I' \left( T_{11.1}, 7.3 \right) \right] \quad \text{otherwise}
   \]

The scheme attempts to account for the effects of multiple layer clouds within the pixel, whilst also remaining relatively simple to implement. Figure 2 provides a graphical illustration of the interpolation scheme for a pixel containing SO$_2$.

The difficulty in estimating the background radiance lies in the non-linear temperature response from a pixel of mixed composition (i.e. clear-sky and clouds at multiple levels). Ideally the scheme should be designed to first estimate the fractional coverage of cloud within the pixel, together with an estimate of the clouds’ height or radiating temperature. Then, under less restrictive assumptions the radiance at 7.3 µm is estimated from clear and cloud-sky radiances separately and combined by weighting the radiances with the fractional cover estimate. Further accuracy could be gained by employing a radiative transfer model with forward calculations and utilising the full suite of HIRS/2 channels. Such improvements are beyond the scope of this paper, and we readily concede that this retrieval scheme suffers limitations.

3.2 The Effects of the HIRS Filter Function

The HIRS filter functions cover approximately 0.44 µm and the response is not constant across the band. Pierlusi

![Figure 2. Radiance vs. Wavelength for a pixel which contains SO₂. The curves are blackbody radiances at the temperatures found by inverting the Planck function at 6.7 µm and 11.1 µm. The + shows the adjusted linearly interpolated radiance at 7.3 µm, the open circle shows radiance at 7.3 µm derived from Eq. 5, and the solid circles show the measured radiances. The difference between the observed 7.3 µm temperature and the estimated 7.3 µm temperature is the temperature anomaly.](image-url)
et al. [1984] provide spectral band parameters for SO$_2$ that can be used to assess the effect of the HIRS filter response on the transmittance. The analysis indicates that the effect is small but important. The difference between assuming a monochromatic transmittance function and doing an integration across the HIRS filter function varies with absorber amount and with the height of the SO$_2$ cloud. The transmittance difference was found to be less than 0.05 over the whole parameter range and peaked between 10–20 m atm-cm. Above 50 m atm-cm the transmittance difference was less than 0.02. The ESFT scheme employed here accounts for the effects of the HIRS filter functions.

3.3 The Effects of the Surface

The analysis indicates that the surface radiance has only a small effect on the retrieval. This lack of dependence arises through the use of radiances from different channels for the same pixel when estimating the unperturbed radiance at 7.3 µm, and from the fact that this channel is almost opaque, except for very dry atmospheres. For optically thin veils of SO$_2^3$, the sensitivity of the scheme will prohibit accurate retrievals. A test is used to limit the retrieval to cases where,

$$T_{11.1} < 295K,$$

where $T_{11.1}$ is the (Planck) brightness temperature sensed in the 11.1 µm channel at the top of the atmosphere.

3.4 The Effects of Water Vapour

Residual water vapour absorption may cause anomalies in the retrieval. If there is significant water vapour absorption within the SO$_2$ cloud then this will also show up as extra absorption in the 6.7 µm channel. This channel is used in the linear interpolation scheme so the algorithm will compensate for the effect of in-cloud water vapour. For cases where there are very high water vapour amounts lying just below the SO$_2$ cloud it is possible that the retrieval will produce anomalous results. The effect will be worse if the SO$_2$ cloud is thin—this sets a limit on the lowest SO$_2$ amount that can be retrieved. There may also be situations where the vertical water vapour distribution causes the linear interpolation scheme to break-down, however these are unlikely to be common situations as they require unusual atmospheric conditions. Finally, SO$_2$ lying below significant water vapour will not produce a useful signal because the 7.3 µm channel is sensitive to water vapour. This puts a constraint on the lowest level in the atmosphere for which SO$_2$ can be measured. A rule-of-thumb estimate for this is 3 km, which is the mean water vapour scale height [Randel et al., 1996]. A test is employed to eliminate water vapour anomalies. To pass the test, pixels must satisfy the condition:

$$T_{11.1} > T_{6.7}.$$

Essentially this test regards the brightness temperature in the window channel (11.1 µm) as always being larger than the brightness temperature in the water vapour channel (6.7 µm). There are occasions when this may not be true (e.g. strong inversions). The test eliminates very few pixels from the retrieval process.

A further test checks that the brightness temperature at 8.2 µm (or 12.5 µm for NOAA-11 and beyond) is larger than the brightness temperature at 11.1 µm, when the 11.1 µm brightness temperature is low (250 K is used). This eliminates some volcanic ash signals and abnormal water vapour and cloud signals. The test is conservative.

3.5 The Effects of Clouds

Water/ice clouds lying above the SO$_2$ cloud will mask SO$_2$ absorption and consequently the scheme will not ‘see’ the SO$_2$. This can occur for very high, thick (cold) cloud, particularly in the tropics. A test has been devised to limit the retrieval to cases where,

$$T_{11.1} > 200K.$$

This test also doubles to eliminate problems arising from thin SO$_2$ veils overlying very cold surfaces (e.g. ice covered surfaces in winter), which suffer from the same constraints as described in 3.3.

A second test is employed to remove the effects of poor calibration, pixel misalignment and some abnormal conditions, notably very high water vapour loadings and strong cirrus cloud signals. The test is:

$$T_{8.2} - T_{11.1} < -10K.$$

For satellites after NOAA-10 the test is:

$$T_{12.5} - T_{11.1} < -10K.$$

The thresholds used in these tests were determined through extensive data analysis and trial-and-error. A better approach would be to use an objective cloud detection scheme, such as English et al. [1999]. Work is in progress to
include such a scheme. The current tests are preliminary and need refinement.

3.6 Data Integrity Tests

The TOVS data processing is complex and occasionally suspect data are delivered into the processed HIRS/2 data files. There appears to be no systematic problem with the data processing, rather anomalies appear to occur in an unpredictable, almost random manner. To eliminate spurious data a simple spike test has been employed. This takes the form of calculating the mean of 8 neighbouring pixels and testing the pixel being processed against the mean. If the pixel value lies well outside the mean the retrieval halts for this pixel. There is a small cost of losing valid data. That is, having a valid pixel with an anomalously high valid value in the neighbourhood of low values, being eliminated in the process.

3.7 Sensitivity to Cloud Height

The conversion of $\Delta T$ measurements into transmittance is relatively straightforward via (3), and the transmittance model (1) allows an estimate of the absorber amount $u$. However, this conversion has a dependence on pressure and temperature of the absorber (through the pressure and temperature dependence of the ESFT coefficients $a_i$ and $k_i$); in other words it depends on the location of the SO2 in the vertical. The dependence on pressure is stronger than that on temperature. Thus, the retrieval requires a height estimate for the SO2. Figure 3 illustrates the variation of SO2 absorber amount ($u$) with temperature anomaly ($\Delta T$) for different cloud heights.

4. ERROR ANALYSIS

Several sources of error have been identified in the retrieval of SO2 from HIRS/2 infrared measurements. We class these errors into two types: those that contribute to the accuracy of the retrieval and those that limit the operation of the retrieval. The first type of errors (Type I) include as major sources:

- Inherent accuracy of the measurements—the NE\Delta T,
- Errors in estimating the background radiance,
- Transmittance modelling errors,
- Error in SO2 cloud height and temperature estimation.

The Noise Equivalent Temperature Difference (NE\Delta T) of HIRS/2 channels 8, 11 and 12 are ~0.5 K. The background radiance errors are difficult to estimate theoretically, but can be estimated by analysis of large amounts of HIRS/2 data when no SO2 clouds are present. These analyses suggest an rms error of ±1 K. Transmittance modelling errors include SO2 band parameter errors, errors due to misfit in the ESFT procedure, errors in the filter response specification, and errors in the relation between SO2 transmittance and 7.3 µm temperature differences. The largest of these four sources of error is the latter and we have estimated this to be ±1 K, based on radiative transfer simulations. Combining these errors and assuming that they are independent, the Type I error is:

$$\delta T_i = \pm 1.5 K.$$  

This can be easily assessed by conversion into an error in SO2 amount by using the sensitivity of the double-exponential model [Pierluisi et al., 1984]. Figure 4 illustrates Type I error in retrieved SO2 for measurement and modelling errors and two cloud heights.

For relatively low clouds, SO2 amounts of < 5 D.U. (1 D.U.=1 m atm-cm) are subject to large error and are probably indistinguishable from the noise (unless spatial coherence is evident). For higher clouds this lower thresh-
old on the error increases to ~20 D.U. In both cases errors of 5–10% are evident throughout the range of expected absorber amounts. The modelling errors dominate measurement errors—which is encouraging as improvements in the model are possible whilst improving measurement errors are only possible by some form of averaging (e.g. spatial and temporal).

The second type of errors (Type II) include:

- Infrared contrast,
- Model sensitivity,
- Anomalous atmospheric conditions,
- Spectroscopic considerations.

Type II errors are systematic and result from basic violations of the measurement and/or atmospheric assumptions or from lack of knowledge of the physical processes. Because the technique described here involves an infrared atmospheric emission measurement, the ability to retrieve SO$_2$ relies on the difference in temperature between the emitting source (the SO$_2$) and the background temperature (due to water vapour or clouds). The lack of ‘infrared contrast’ fundamentally restricts the technique. Thus if the SO$_2$ cloud is at the same temperature as the water vapour/cloud/ atmosphere below, it is not possible to reliably retrieve the SO$_2$ amount regardless of its magnitude. Although on average the infrared contrast is adequate, there are persistent conditions where retrieval accuracy is limited. These include geographic locations where the surface below is as cold as the SO$_2$ cloud itself, for example, over ice-covered Antarctica and Greenland and climatic regions where the SO$_2$ cloud lies above the tops of high, thick clouds, such as tropical cumulonimbus clouds. Inspection of (2) and (3) shows that $\Delta T \rightarrow 0$, as the infrared contrast decreases ($B_s - I_a$), regardless of the amount of SO$_2$ ($t_s$).

Another drawback of the infrared measurement arises through lack of sensitivity of the model. There are two limiting cases for which SO$_2$ retrieval is problematic. For very high SO$_2$ amounts, inspection of (2) shows that as $t_s \rightarrow 0$, the measured radiance arises solely from the SO$_2$ cloud and consequently the infrared contrast is small. The transmittance model suggests that absorber amounts $>\sim 900$ D.U. cause saturation. Using the optimal parameters for $\alpha$ and $\beta$ in (3) with $t_s = 0$, suggests $\Delta T = -40$ K. For very low SO$_2$ amounts, $t_s \rightarrow 1$ and (3) gives $\Delta T = \alpha$. This case can be investigated further by assuming that values with $\Delta T$ below $\alpha - |\delta T_1|$ are at the detection limit. In this case the model gives 3.7 D.U. for a cloud at a height of $\sim 8$ km. In practice smaller amounts may be detected by assessing the spatial coherence of the data or by averaging pixels.

Atmospheric conditions for which the basic assumptions of the model are violated include cases where water vapour and/or cloud lies above the SO$_2$ cloud or when significant water vapour is collocated with the SO$_2$. Unusual inversions in the temperature and/or water vapour profile may also cause problems with the retrieval. All of these anomalous conditions do occur, but experience with processing the data suggests that they are not common. Perhaps the main area of concern is the occurrence of water vapour coexisting with SO$_2$ and/or occurring in large enough quantities to cause significant absorption across the 7.3 $\mu$m band. This problem is somewhat alleviated because the retrieval scheme makes use of radiance measurements at 6.7 $\mu$m, which changes as the mid-tropospheric water vapour changes in a similar fashion to the radiance change at 7.3 $\mu$m. Thus the scheme compensates for extra water vapour associated with the SO$_2$ cloud. Demonstrating unequivocally that water vapour is not a seri-

![Figure 4. (a) Measurement and modelling errors as a function of absorber amount for a plume at 5 km height and low abundance. (b) Measurement and modelling errors as a function of absorber amount for a plume at 35 km height and high abundance.](image-url)
ous problem will need to wait until routine high spectral resolution data are available across the 7.3 $\mu$m band (e.g. from AIRS data—see Chahine et al., [2003]).

5. EXAMPLES

5.1 Pinatubo

Quantitative use of HIRS/2 data to estimate $SO_2$ has not been made previously and it is therefore necessary to demonstrate what is possible using examples from well-known cases of volcanic $SO_2$ clouds. One such case is that of the June 1991 Pinatubo eruptions which are generally regarded to have injected ~15–19 Tg of $SO_2$ into the stratosphere [Self et al., 1996]. Such a large $SO_2$ anomaly should be readily detectable in the HIRS/2 data.

The Pinatubo $SO_2$ cloud was measured by the TOMS instrument and also by several other airborne and satellite-borne sensors [McCormick et al., 1995; Self et al., 1996]. During 1991 three HIRS/2 instruments were flying (see Table 2)–on NOAA-10, NOAA-11 and NOAA-12. These satellites had different node crossing times and, since HIRS/2 is an infrared sensor both day and night coverage was possible. Plate 1 shows a time sequence of $SO_2$ retrievals made from the three HIRS/2 instruments and Table 4 provides a comparison of total masses (determined in the same way as in Krueger et al., 1995, Eq. 15) retrieved from combined HIRS/2 measurements (3 instruments) with TOMS for several days. Generally, the agreement is good: HIRS/2-TOMS=+1.0 Tg, $\sigma = \pm 1.2$ Tg.

5.2 Cerro Hudson

In August 1991, Cerro Hudson erupted sending a large amount (> 1 Tg) of $SO_2$ into the upper troposphere and lower stratosphere. HIRS/2 retrievals have successfully tracked the $SO_2$ cloud for nearly 3 weeks as it circled the southern hemisphere at latitudes south of 30 °S. Plate 2 shows a montage of $SO_2$ retrievals obtained over a 9-day period as the cloud encircled the hemisphere. Numerical modelling of the cloud transport and TOMS data verify that the cloud passed over southern Australia and the Tasman sea, causing several aviation encounters [Doiron et al., 1991; Barton et al., 1992].

The HIRS/2 retrievals gave very similar results to those obtained from TOMS and because of the greater temporal sampling offer new opportunities to estimate the decay rate of stratospheric $SO_2$ and permit new studies of the conversion rate of $SO_2$ to $H_2SO_4$. Both of these parameters are important to the chemistry of atmospheric $SO_2$ [McKeen et al., 1984]. Diurnal effects on $SO_2$ and the relation between $SO_2$, $H_2SO_4$ and $O_3$ (using the HIRS/2 9.7, $\mu$m channel) are also possible.

5.3 Ruapehu

The examples given so far can be readily verified against the TOMS data. During 1996 no TOMS instruments were flying and consequently little is known about the $SO_2$ stratospheric loading during that year. However, there were several volcanic eruptions reported, some of which are likely to have injected $SO_2$ into the upper troposphere/lower stratosphere.

Between June and August 1996, Mt Ruapehu on the North island of New Zealand erupted several times injecting ash and $SO_2$ into the atmosphere. Satellite tracking of the Ruapehu plumes using AVHRR data suggested that the ash content of the eruption clouds was significant [Prata and Grant, 2001]. Ground-based monitoring of the emissions also suggested that there was significant $SO_2$ degassing. The first satellite measurements of the $SO_2$ emissions from Ruapehu are presented here. Results are shown in Plate 3 for retrievals from the NOAA-12 and NOAA-14 HIRS/2 instruments. Maximum $SO_2$ amounts did not exceed 100 D.U.; the average amounts were ~20–30 D.U. The total mass loadings were less than 0.05 Tg-values of 0.03, 0.02, 0.02, and 0.01 Tg were found for the 17 June, 1996 eruption.

5.4 Hekla–Trajectories

A fourth example of detection and monitoring of relatively small (~< 1 Tg), stratospheric $SO_2$ clouds from eruptions

<table>
<thead>
<tr>
<th>Date</th>
<th>HIRS/2</th>
<th>TOMS</th>
<th>HIRS/2</th>
<th>TOMS</th>
</tr>
</thead>
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<td>16-June-1991</td>
<td>15.2</td>
<td>15.5</td>
<td>±1.8</td>
<td>±1.2</td>
</tr>
<tr>
<td>17-June-1991</td>
<td>18.7</td>
<td>18.5</td>
<td>±2.2</td>
<td>±1.6</td>
</tr>
<tr>
<td>18-June-1991</td>
<td>16.5</td>
<td>16.0</td>
<td>±1.2</td>
<td>±1.1</td>
</tr>
<tr>
<td>19-June-1991</td>
<td>15.9</td>
<td>14.0</td>
<td>±1.1</td>
<td>±0.9</td>
</tr>
<tr>
<td>20-June-1991</td>
<td>17.4</td>
<td>14.5</td>
<td>±1.1</td>
<td>±0.9</td>
</tr>
<tr>
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<td>14.0</td>
<td>±0.9</td>
<td>±1.7</td>
</tr>
<tr>
<td>22-June-1991</td>
<td>16.9</td>
<td>14.0</td>
<td>±1.6</td>
<td>±1.7</td>
</tr>
<tr>
<td>23-June-1991</td>
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<td>14.0</td>
<td>±1.8</td>
<td>±1.7</td>
</tr>
<tr>
<td>24-June-1991</td>
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<td>14.0</td>
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<td>±1.7</td>
</tr>
<tr>
<td>25-June-1991</td>
<td>11.5</td>
<td>14.0</td>
<td>±1.7</td>
<td>±1.7</td>
</tr>
</tbody>
</table>
Plate 1. Sequences of SO$_2$ column abundance (in m atm-cm) from the 15 June, 1991 Pinatubo eruption retrieved from three HIRS/2 instruments.
Plate 2. Montage of the SO$_2$ plume erupted from Cerro Hudson showing how it encircled the southern hemisphere in about 9 days, during August 1991.
of Hekla volcano, Iceland is provided. These SO$_2$ clouds typically contain SO$_2$ amounts of 50 D.U. or less and show that robust detection at the lower limit of detection is possible. We illustrate these retrievals in Figure 5 (a) and Figure 5 (b) by showing the trajectory of the maximum SO$_2$ amount retrieved during each orbital pass of the satellite. Results are shown for the 1980 and 1991 Hekla eruptions, and demonstrate how these measurements could be used to validate dispersion/trajectory modelling analyses.

5.5 Potential Climate Studies

The importance of sulphur-rich volcanic eruptions to climate has been discussed by Rampino and Self [1984]. The SO$_2$ retrievals from HIRS/2 are global, have relatively high spatial resolution (up to $18 \times 18$ km$^2$ at nadir) and temporal resolution (up to 6 times per day from 3 satellites) and span nearly 24 years. This makes this data set useful for climate studies. To date we have made global SO$_2$ retrievals for all three HIRS/2 instruments operating during 1991 and have begun analyzing data for other years. The global imprint of SO$_2$ in the upper troposphere/lower stratosphere during 1991 is shown in Plate 4.

The top panels in the plate show the zonally averaged SO$_2$ amount, while the lower panels show the zonally integrated SO$_2$ amount. Intercomparison of the panels provides an estimate of the consistency of the retrievals from two different HIRS/2 instruments. During 1991 there were two climatically significant eruptions: Pinatubo and Cerro Hudson. Pinatubo’s SO$_2$ influence lasted more than 2 months and penetrated well into northern hemisphere midlatitudes. Cerro Hudson’s influence was largely restricted to southern hemisphere mid-to-high latitudes and lasted 2–3 weeks. The radiative impact of SO$_2$ has been studied by Gerstell et al. [1995] who recommended that general circulation models should include the radiative forcing due to SO$_2$ gas as well as H$_2$SO$_4$ and volcanic ash. Pudykiewicz and Dastoor [1995] simulated the global distribution of sulfate aerosols produced by Pinatubo and found good agreement with aerosol optical depth measurements. These authors introduced the concept of a virtual source—an analytic function that describes the spatial and temporal release of SO$_2$ into the stratosphere. The analyses presented here could be used as initial conditions for a model that includes the radiative effects of SO$_2$ and replaces the need for an analytic source function.

6. CONCLUSIONS

The retrieval of SO$_2$ from HIRS/2 data provides a new global, long-term data set for investigating and quantifying the stratospheric SO$_2$ budget. These new measurements complement existing TOMS SO$_2$ data and permit intercomparisons of two independent SO$_2$ estimates. The HIRS/2 retrieval scheme works best when the SO$_2$ is separated from water vapour and lies above it—the scheme provides a natural filter for detecting SO$_2$ that reaches the upper troposphere/lower stratosphere, where its climatic implications are greatest. The error analysis suggests that accuracies are in the range 10–20% for SO$_2$ column amounts in the range 10–800 m atm-cm. Fundamental difficulties with using infrared measurements prevent making accurate retrievals below and above these limits. Improvements can be made in the retrieval scheme, most notably by using more detailed...
Plate 4. (a) Zonally averaged SO$_2$ column abundance (m atm-cm) as a function of time and latitude during 1991, derived from the NOAA-10 HIRS/2. (b) Zonally integrated SO$_2$ column abundance (m atm-cm) as a function of time and latitude during 1991, derived from the NOAA-10 HIRS/2. (c) Zonally averaged SO$_2$ column abundance (m atm-cm) as a function of time and latitude during 1991, derived from the NOAA-11 HIRS/2. (d) Zonally integrated SO$_2$ column abundance (m atm-cm) as a function of time and latitude during 1991, derived from the NOAA-11 HIRS/2.
radiative transfer modelling, an improved cloud detection scheme, and better use of the multispectral measurements available with the HIRS/2 instrument. The scheme presented here is fast and ideally suited to operational processing of global data in near real-time.

Further analysis of the HIRS/2 data will permit retrieval of volcanic ash, H$_2$SO$_4$ (cf. Ackerman and Strabala [1996] and Baran and Foot [1994]) as well as SO$_2$, providing a more comprehensive set of data for understanding the climatic influence of volcanic eruptions. Use of the 9.7 µm HIRS/2 channel in combination with the SO$_2$ retrievals may also provide new information on the effects of volcanic aerosols on ozone depletion.

The retrieval scheme is not restricted to HIRS/2 data-any instrument with channels near 6.7, 7.3 and 11 µm is amenable to this technique. The MODIS instruments on board Terra and Aqua can be used to derive SO$_2$ [see Rose et al., 2003 this volume] as can the new multispectral imager-SEVIRI, on board the Meteosat Second Generation platform. Currently the AIRS instrument on board the Aqua platform can be used to derive SO$_2$ by exploiting the 7.3 µm feature, with the strong possibility of using the high-spectral content and the narrow channels (up to 2378) to probe the lower troposphere and derive vertical profile information. Perhaps more exciting is the possibility of using the high-spectral resolution data from the Geostationary Infrared Fourier-Transform Spectrometer (GIFTS), which will provide high spatial, temporal and spectral resolution data capable, in principle, of giving SO$_2$ concentrations at several levels in the upper troposphere/lower stratosphere. To date we have processed only a small part of the considerable global archive of HIRS/2 data. In due course we hope to complete the processing back to the TIROS-N HIRS/2 (1978) and provide a real-time processor for users with access to direct broadcast NOAA data. An operational HIRS/2 SO$_2$ product would be useful for aviation hazard warning as well as for studying local and global effects of SO$_2$ on climate.

Acknowledgments. The authors are grateful to Darren Jackson and John Bates both at the NOAA ARL for processing the level 1a TOVS archive and providing the level 1b radiance data. Three anonymous reviewers provided constructive advice on ways to improve this manuscript and we are most grateful to them.

APPENDIX 1: LIST OF ACRONYMS

AIRS—Atmospheric InfraRed Sounder
ASTER—Advanced Spaceborne Thermal Emission and Reflection radiometer

APPENDIX 2: RADIATIVE TRANSFER MODEL

The model assumed for analysis of HIRS data has a layer of SO$_2$ embedded in an otherwise clear atmosphere between heights $z = a$ and $z = b$. Variations in the density of SO$_2$ and its specific absorption coefficient are assumed to be negligible throughout the layer. The optical thickness from height $z$ to space along a vertical ray is

$$l(z) = \begin{cases} l_a(z) & \text{if } b \leq z \\ l_a(z) + \alpha_s(b - z) & \text{if } a \leq z \leq b \\ l_a(z) + \alpha_s(b - a) & \text{if } z \leq a \end{cases}$$

Here $l_a(z)$ is the optical path from height $z$ to space of an atmosphere without SO$_2$, and $\alpha_s$ is the volume absorption coefficient of SO$_2$. The corresponding transmittances from height $z$ to space are

$$t(z) = \begin{cases} t_a(z) & \text{if } b \leq z \\ t_a(z) \exp(-\alpha_s(b - z)) & \text{if } a \leq z \leq b \\ t_a(z) \exp(-\alpha_s(b - a)) & \text{if } z \leq a \end{cases}$$

where $t_a(z)$ is the transmittance of an atmosphere without SO$_2$. We let $\tau_s$ and $t_s$ denote the optical thickness and transmittance of the SO$_2$ column, defined by

$$\tau_s = \alpha_s(b - a) \quad \text{and} \quad t_s = \exp(-\tau_s). \quad (9)$$

The SO$_2$ layer attenuates the radiance emitted by the atmosphere below, while within the SO$_2$ layer, where both the atmosphere and SO$_2$ are active, absorption by each gas modifies the radiance emitted by the other. The radiance $I$ emitted to space consists of four terms,

$$I = t_s I_a + (1 - t_s) I_a + I_{as} + I_{ss}. \quad (10)$$
The first represents the radiance \( I_a \) emitted to space by an atmosphere free of SO\(_2\),

\[
I_a = B_0 t_a (0) + \int_0^\infty dz \alpha_a(z) B(z) t_a(z),
\]

(11)

attenuated by the transmittance \( t_a \) of the SO\(_2\) column. Here \( \alpha_a(z) \) is the volume absorption coefficient of the atmosphere at height \( z \), \( B_0 \) is the Planck emission by the surface, and \( B(z) \) is the Planck emission from the atmosphere at height \( z \) where the temperature is \( T(z) \). The second term is the radiance \( I_a^+ \) emitted by the atmosphere above the SO\(_2\) layer,

\[
I_a^+ = \int_0^\infty dz \alpha_a(z) B(z) t_a(z),
\]

(12)

in this case modified by \( (1 - t_a) \). The third term represents radiance emitted by the atmosphere within the SO\(_2\) layer and modified there by absorption by SO\(_2\),

\[
I_m = \int_b^\infty dz \alpha_a B(z) t_a(z) e^{-\alpha(z)/b-a}.
\]

(13)

The final term represents radiance emitted by SO\(_2\) and subsequently attenuated by absorption in the atmosphere above the point of emission,

\[
I_m = \int_a^b dz \alpha_a B(z) t_a(z) e^{-\alpha(z)/b-a}.
\]

(14)

Our analysis of HIRS data is based on two additional assumptions that simplify these equations.

1. We assume that the SO\(_2\) layer lies above the peak of the weighting function for H\(_2\)O in the 7.3 \( \mu \)m channel, so that the absorption coefficient \( \alpha_a(z) \) is zero for \( z > a \). This is a significant assumption, because H\(_2\)O absorbs strongly at 7.3 \( \mu \)m and the weighting function peaks relatively high in the atmosphere.

2. The SO\(_2\) layer is assumed to be isothermal, and therefore may be characterized by a single temperature \( T_s \). Clearly, this assumption is questionable when the SO\(_2\) layer is very deep.

Justification for these assumptions is pragmatic, in the sense that the technique appears to track SO\(_2\) plumes from major volcanic eruptions, even though we know that frequently the emission of SO\(_2\) is accompanied by large emissions of H\(_2\)O.

With these assumptions, the formulae simplify. Because \( \alpha_a(z) \) is zero for \( z > a \), the second and third terms in equation (10) are zero, while the fourth term reduces to

\[
I_m + \int_a^b dz \alpha_a(z) B(z) t_a(z) e^{-\alpha(z)/b-a} = (1-t_a) B_s.
\]

(15)

where \( B_s \) denotes the Planck function at the temperature \( T_s \) of the SO\(_2\) cloud. Thus,

\[
I = t_a I_a^+ + (1-t_a) B_s,
\]

(16)

which we also write in the forms

\[
I - I_a^+ = (1-t_a)(B_s - I_a^+) \quad \text{and} \quad e^{\alpha(z)/b-a} = \frac{B_s - I}{B_s - I_a^+}.
\]

Our aim is to estimate \( \tau_a \). The data will be observations of the radiance \( I \) emitted to space in the HIRS 7.3 \( \mu \)m channel and estimates of \( B_s \) and \( I_a^+ \) derived from other HIRS/2 channels insensitive to SO\(_2\).

NOTES

1. Profiles can also be retrieved but this requires much higher spectral resolution.

2. In keeping with previous practice (e.g. Krueger et al. [1995]), we use the units milli atmosphere centimetre (m atm-cm) for absorber amount. 1 m atm-cm = 1 D.U. (Dobson Unit) or in S.I. units 1 atm-cm = 0.446157 mol m\(^{-2}\).

3. An infrared (7.3 \( \mu \)m) optical depth of less than 0.05 is equivalent to \( \varepsilon \) K difference between the background atmosphere and the SO\(_2\) layer.

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D. M. O’Brien, CSIRO Atmospheric Research, PB1, Aspendale, Victoria 3195, Australia, Email: Denis.O’Brien@csiro.au

A. J. Prata, CSIRO Atmospheric Research, PB1, Aspendale, Victoria 3195, Australia, Email: Fred.Prata@csiro.au

W. I. Rose, Geological Engineering and Sciences, Michigan Technological University, Houghton, Michigan, USA, Email: raman@mtu.edu

S. Self, Department of Earth Sciences, The Open University, Walton Hall, Milton Keynes, MK7 6 AASA, United Kingdom, Email:Stephen.Self@open.ac.uk.