# Retrieval of atmospheric sulfur dioxide and nitric acid using the Atmospheric Infrared Sounder (AIRS)

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# ABSTRACT

The Atmospheric Infrared Sounder (AIRS) flying on NASA's EOS-AQUA platform has channels sensitive to both sulfur dioxide (SO2) and nitric acid (HNO3). We have developed a simple regression retrieval for both of these gases that illustrates the potential for AIRS, and other hyperspectral sounders, to retrieve these two gases. We have cross-validated the SO2 retrievals to those from the OMI instrument flying on the EOS AURA platform. Similarly, we have cross-validated the HNO3 retrievals with limb retrievals of HNO3 by from MLS instrument, also flying on the AURA platform.

Keywords: remote sensing, minor gases, sulfur dioxide, nitric acid, atmospheric retrievals

# **1. INTRODUCTION**

The Atmospheric Infrared Sounder (AIRS) is a high spectral resolution grating spectrometer on board the NASA's EOS Aqua satellite platform, and has been in near continuous operation since September 2002. The sun-synchronous orbit of Aqua combined with the nadir viewing cross-track scanning of AIRS provides observations of the majority of Earth's surface twice each day, once during the day and again ~12 hours later at night. AIRS has 2378 spectral channels spanning 650-1615 and 2180-2670 cm<sup>-1</sup> with a resolution of approximately  $\nu/1200$ . A number of these channels are sensitive to the atmospheric concentration of trace gases SO2 (sulfur dioxide) and HNO3 (nitric acid). See a recent review article on AIRS<sup>1</sup> and references therein for an overview of the AIRS instrument and retrieval products.

The "version 5" AIRS operational retrieval software package used by the Goddard Earth Sciences Data Information Services Center (GES DISC) to process AIRS observations and produce data products (temperature, water vapor, ozone) does not include retrievals of either SO2 or HNO3. This paper gives an overview of some preliminary work to explore the potential of AIRS to retrieve these trace gases. A relatively simple retrieval technique is used, with encouraging results.

#### **2. SULFUR DIOXIDE**

Under ordinary conditions, SO2 is present in the atmosphere at background levels on the order of 0.1 Dobson Unit (DU). A Dobson Unit is the thickness in hundredth of a millimeter of a gas compressed to a pressure of 1 atmosphere at 0 C. This low background level is too low for AIRS to detect, but volcanic eruptions often release large amounts of SO2 into the atmosphere creating SO2 plumes of tens or hundreds of DU that can travel long distances before diffusing away. AIRS has some ability to detect and measure these SO2 plumes when the gas concentration is well above typical background levels. We have previously detected SO2 with AIRS from a major eruption of Mt. Etna,<sup>2</sup> using a preliminary retrieval algorithm. In that work we also showed that AIRS can detect and track the evolution of a volcanic ash cloud if it exists with sufficient optical depth. Note that the spectral signatures of the ash and SO2 clouds do not overlap, allowing both to be retrieved without interference from the other.

AIRS channels with some sensitivity to the concentration of SO2 present in the observation are shown in the top panel of Figure 1, which shows a simulated AIRS clear sky brightness temperature (BT) spectra for the 950-1450 cm<sup>-1</sup> region for three SO2 profiles of 0.1, 100, and 1000 DU. The bottom panel shows the change in BT due to the change in SO2. Each year there are usually a few eruptions somewhere on the globe with SO2 plumes on the order of 100 DU, while eruptions on the order of 1000 DU are very rare (Pinatubo in 1991 was the most recent).

As can be seen from Fig. 1, the AIRS channels most sensitive to changing SO2 are located in the 1370 cm<sup>-1</sup> region. For a typical background level SO2 profile, the atmospheric absorption there is dominated by water vapor, but also with significant contributions by CH4 and N2O. With a careful selection of channels the CH4 and N2O can be avoided, but the water vapor affects all the 1370 cm<sup>-1</sup> region channels. The water vapor absorption here blocks most of the upwelling



Figure 1. Sensitivity of AIRS to SO2.

radiance originating at the surface and the lower troposphere, thereby preventing these channels from detecting low altitude SO2 plumes. The  $1100 \text{ cm}^{-1}$  region channels can see to the surface, but they can only detect SO2 if it is on the order of 100 DU or more.

Our SO2 (and HNO3) retrieval method is based on regression. We started by selecting two sets of AIRS channels: one set with strong SO2 absorption, and another set with weak SO2 but with otherwise similar absorption characteristics as the strong SO2 channels. That is, we chose our channels such that with nominal background level SO2 the two sets of channels respond to the atmospheric state in similar ways and AIRS observes similar BTs. With such a channel set it may be possible to use the BT of "weak" channels to predict the BT of the "strong" channels under the assumption of nominal background level SO2. We do a simple regression of the form

$$A \times X = B \tag{1}$$

where A is a matrix of "weak" channel observed BTs (the predictor matrix), X are the coefficients to be determined by regression, and B are the "strong" channel BTs. The regression was trained on simulated spectra (including random noise), and once the coefficients X are determined the above equation can be used to predict the "strong" BT using real observed data.

If the actual SO2 present in the observation is substantially higher than the nominal level, then the BT predicted for the "strong" channels will differ from the actual measured BT. The size of this  $\Delta$ BT will depend on the amount of excess SO2. Our AIRS fast model<sup>3,4</sup> allows for variable SO2, so if we have an estimate of the atmospheric temperature and water vapor profiles, we can compute the expected delta BT for a given amount of SO2. Using an iterative approach we can then estimate the amount of SO2 present in the observation.

Figure 2 shows the AIRS channels we selected for use in our SO2 retrieval. The top panel shows a blowup of the BT spectra, while the bottom panel shows AIRS sensitivity to changes in SO2 as well as CH4 and N2O. The open circles are channels sensitive to SO2, while the solid dots are the SO2 insensitive channels. Note the avoidance of channels strongly sensitive to CH4 and N2O in the bottom panel. The effect of increasing the SO2 from 0.1 to 1 DU shown in the bottom panel changes the AIRS BT by at most 0.15 K, which is similar to the AIRS noise level. Thus 1 DU is a rough estimate of the lower limit of AIRS sensitivity to SO2.

Another consideration for AIRS SO2 retrievals are cloud effects. Besides the usual cloud cover found across the globe, volcanic eruptions releasing large amounts of SO2 also tend to release substantial amounts of water vapor and sometimes also ash. As the volcanic steam rises, it will cool and form clouds. These volcanic clouds often reach high into the troposphere, sometimes all the way to the tropopause. AIRS will not be able to detect the SO2 below these clouds.



Figure 2. AIRS channels selected for SO2 retrievals. Top panel is an AIRS simulated spectrum, the bottom panel shows the sensitivity of this spectrum to SO2, CH4, and N2O.





AIRS has very limited ability to determine the vertical location of an SO2 plume. If we can clearly see an SO2 plume using the  $1370 \text{ cm}^{-1}$  channels then we know a significant amount of SO2 must have reached the upper troposphere, but we do not know the shape of the profile. That is, we can not tell if the SO2 plume extends all the way from the tropopause down to the surface, or if it is restricted to some sort of slab in the upper troposphere. In general, AIRS is only sensing a portion of the vertical column, and so we must assume a vertical profile shape when estimating the total column.

Figure 3 shows our AIRS retrieval of an SO2 plume near Manam in New Guinea on 28 January 2005. Also shown for comparison is the SO2 retrieval from the Ozone Monitoring Instrument (OMI).<sup>5</sup> For this comparison the AIRS retrieval assumed the SO2 resided in a slab confined to approximately 200-350 mb. On the whole, the retrieved SO2 agrees well with OMI, but AIRS SO2 column values are dependent on the assumed vertical location of the SO2 plume. Moving the assumed location 100-200 mb gives a retrieval that looks almost exactly the same except the column amounts are halved!



Figure 4. Sensitivity of AIRS to HNO3. Top panel is a simulated AIRS spectrum. The bottom panel is the change in BT for a 50% increase in the HNO3 column.

In some case, clouds can be used to help provide information about the vertical location of the SO2 plume. If the field of view of an AIRS observation is completely filled by a cloud, then AIRS channels that ordinarily see to the surface in clear sky conditions will instead see a BT corresponding to the temperature at the top of the cloud. Combined with knowledge of temperature profile, this can be used to estimate the vertical location of cloud top. If AIRS observes enhanced SO2 under these conditions, then we know the SO2 plume extends above the cloud top. The tropopause tends to act as a cap on mixing of air from the tropopause and stratosphere, so we can also assume the SO2 plume will rarely extend above the tropopause.

## **3. NITRIC ACID**

Our retrieval method for HNO3 is similar in concept to that discussed for SO2 above, with some important differences. Unlike SO2 which is subject to huge variations in concentration due to volcanos, the range of variation of HNO3 is much smaller, and we want to measure HNO3 amounts less the nominal mean as well values greater. Also most of the variations in HNO3 column are found in the stratosphere and not the troposphere as was the case for SO2. The nominal global average of HNO3 is about 0.4 DU, and the range of variation is roughly 0.2 to 5 times the nominal mean.

The top panel of Fig. 4 shows a typical AIRS spectrum for the 650-1620 cm<sup>-1</sup> channels, and the bottom panel shows the delta BT resulting from increasing HNO3 by 50%. There are two clumps of AIRS channels with strong sensitivity to HNO3: one in the 880 cm<sup>-1</sup> window region, and the other in the 1330 cm<sup>-1</sup> region.

The  $1330 \text{ cm}^{-1}$  channels are dominated by water absorption, but there is also significant CH4 and N2O absorption in some channels which our retrieval avoided. The water absorption blocks most radiation from the surface and lower troposphere for typical tropical and mid-latitude profiles, but over the poles where the air is usually very dry and the surface altitude is often high, these lower troposphere/surface radiance contributions can become non-negligible.

The 880 cm<sup>-1</sup> channels are a little noisier than 1330 cm<sup>-1</sup>, but much less affected by other absorbing gases. Surface and lower tropospheric radiance contributions dominate total radiance observed in the 880 cm<sup>-1</sup> channels, so this makes the channels more sensitive to surface modeling errors and the effects of low altitude clouds than the 1330 cm<sup>-1</sup> channels. For these reasons we used the 1330 cm<sup>-1</sup> channels for our retrieval.

Due to the relative small range in delta BT due to HNO3 variability, our retrieval requires an accurate atmospheric temperature and water vapor profile, and cloud effects need to be accounted for. We therefore chose to do our HNO3 retrieval using the AIRS Level 2 support products profile and cloud clear radiances.



Figure 5. Comparison between observed HNO3 from MLS (top panel) and AIRS (bottom panel). Colorbar units are Dobsons. Map scales are latitude (horizontal) and longitude (vertical) in degrees.

Figure 5 shows a comparison of our global AIRS HNO3 retrieval for 22 July 2006 and the version 1.5 HNO3 product of the Microwave Limb Sounder (MLS).<sup>6</sup> The spatial coverage of MLS is much less than AIRS, but MLS HNO3 should be more accurate than AIRS. The agreement of the two retrievals for this particular case is fairly good, but in other cases we see offsets between the two instruments. AIRS retrievals tend to be biased high compared to MLS, and AIRS tends to have more HNO3 over the poles than MLS. In addition there is a small but noticeable limb-darkening effect bleeding into the AIRS retrievals. However, these results are quite encouraging given the simple approaches we have used to date.

#### 4. CONCLUSIONS

We have demonstrated that AIRS has some ability to retrieve quantitative information about volcanic SO2 plumes and global HNO3.

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