Quantifying tropospheric volcanic emissions with AIRS: The 2002 eruption of Mt. Etna (Italy)

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[1] The Atmospheric Infrared Sounder (AIRS) is a hyperspectral IR spectrometer orbiting on the EOS/Aqua spacecraft since May 2002. In late October 2002, AIRS detected lower tropospheric sulfur dioxide and ash emitted by an eruption of Mt. Etna (Italy), in plumes which could be tracked over 1000 km from the volcano into north Africa. We report retrievals of SO_2 and ash column amounts and ash particle size in the Etna plumes. AIRS total SO_2 compares favorably with contemporaneous ground-based correlation spectrometry (COSPEC) measurements. Retrieval of ash cloud parameters in the eruption plumes permits quantitative studies of distal ash fallout. The Etna data demonstrate the potential of AIRS to improve measurements of volcanic SO_2 and ash loading in the troposphere, and to refine our understanding of volcanic cloud composition, structure and evolution. **Citation:** Carn, S. A., L. L. Strow, S. de Souza-Machado, Y. Edmonds, and S. Hannon (2005), Quantifying tropospheric volcanic emissions with AIRS: The 2002 eruption of Mt. Etna (Italy), *Geophys. Res. Lett.*, *32*, L02301, doi:10.1029/2004GL021034.

1. Introduction

[2] Emanations from active volcanoes are significant sources of greenhouse, acid and trace gas species and aerosols. Space-based measurements of these emissions support studies of volcano-atmosphere interactions and comparisons of natural and anthropogenic emission inventories, and enable tracking of airborne ash for aviation hazard mitigation. Monitoring of sulfur dioxide (SO_2) emissions is imperative as the gas correlates robustly with volcanic activity and is a precursor of atmospheric sulfate aerosol. Quantitative sensing of volcanic SO_2 from space was initiated by the ultraviolet Total Ozone Mapping Spectrometer (TOMS) instrument and the TOMS database is the longest extant record of volcanic SO_2 production [*Carn et al.*, 2003], although a similar chronology is expected from infrared HIRS/2-3 data [*Prata et al.*, 2003]. However, the low spectral and spatial resolution of TOMS and HIRS/2-3 bias current long-term records in favor of volcanic clouds in the upper troposphere and above, leaving lower tropospheric emissions to be inferred from sporadic sub-orbital measurements [e.g., *Andres and Kasgnoc*, 1998].

[3] Current IR instruments have enhanced SO_2 and aerosol detection capabilities (e.g., MODIS, ASTER) [e.g., *Corradini et al.*, 2003], but space-based quantification of lower tropospheric volcanic emissions during small eruptions is still challenging as the clouds are less extensive, contain lower concentrations of target species, and have shorter lifetimes than higher-magnitude discharges. Operational ash detection is achieved using geostationary IR satellites (e.g., Washington Volcanic Ash Advisory Center (VAAC: http://www.ssd.noaa.gov/VAAC) but coincident SO_2 production is rarely quantified. The UV GOME and SCIAMACHY instruments have high sensitivity to tropospheric SO_2 [e.g., *Borrell et al.*, 2003] but restricted temporal resolution since they do not provide contiguous coverage.

[4] Here we show that the Atmospheric Infrared Sounder (AIRS) on the EOS/Aqua spacecraft offers the potential for improved quantification of eruptive volcanic emissions in the troposphere. Since late 2002, AIRS has sensed SO_2 or ash emitted by several volcanoes including Reventador (Ecuador), Anatahan (Mariana Islands), Soufriere Hills (Montserrat), Ruang and Bromo (Indonesia), Nyamuragira (D.R. Congo), and Shiveluch (Kamchatka) (e.g., http://toms.umbc.edu). Most of these eruptions produced upper tropospheric or stratospheric clouds also detected by other satellite sensors. A better demonstration of the capabilities of AIRS was permitted by the eruption of Mt. Etna (Italy; ~3300 m altitude) in October 2002, which emitted SO_2 and ash clouds to altitudes of ~6 km. We present AIRS retrievals of SO_2 and ash in Etna's eruption plumes that illustrate the advantages of high spectral resolution for lower tropospheric measurements.

2. The AIRS Instrument on EOS/Aqua

[5] AIRS was launched in a polar orbit (1:30 p.m. ascending node) on EOS/Aqua in May 2002. The instrument is a 2378-channel sounder providing IR coverage from 649–1136, 1217–1613 and 2169–2674 cm^{-1} at high spectral resolution ($\lambda/\Delta\lambda$ = 1200; ~0.5 cm^{-1}) (http://www-airs.jpl.nasa.gov). This includes the strong ν_3 absorption band of SO_2 centered at ~1362 cm^{-1} and the weaker ν_1+ν_3 band at ~2500 cm^{-1}, but only partially covers the ν_1 band centered at ~1160 cm^{-1} (Figure 1). Ash and sulfate aerosol absorption features in the 830–1250 cm^{-1} range are also covered (Figure 1). Spatial resolution is 13.5 km at nadir, and the NEΔT (noise equivalent change in tempera-

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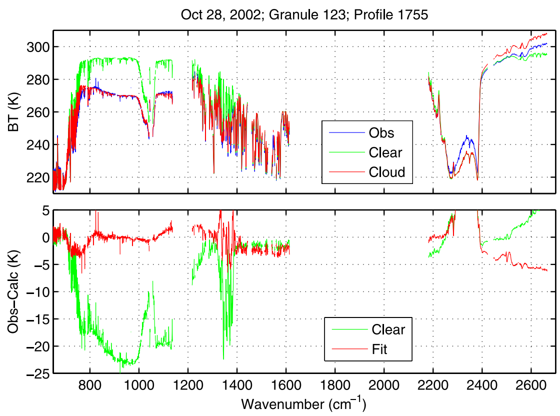

Figure 1. Spectra from the Etna plume on 28 October 2002 at 1218 UT. Upper plot shows brightness temperatures (BTs); showing AIRS data (*Obs*), clear sky spectrum derived from European Center for Medium Range Weather Forecasts (ECMWF) model radiances (*Clear*) and the result after fitting for SO_2 and ash parameters using SARTA (*Cloud*). Lower plot shows clear sky bias (*Clear*) and bias after fitting for SO_2 and ash (*Fit*). Note the SO_2 signal around 1362 cm^{-1}. The ash signal is manifested in a negative slope between 800 and 1000 cm^{-1} in the clear sky bias. Cirrus (ice) clouds show a positive slope in this spectral region.

ture) is ~0.2K from 735–2674 cm^{-1}. In a 24-hour period AIRS nominally observes the complete globe once by day and once by night.

[6] For volcanic SO_2 retrievals use of the ν_3 band is advantageous as it is around an order of magnitude stronger in intensity than the ν_1 band and less affected by interference from aerosol, but the ν_3 band does not lie in an atmospheric window (Figure 1). Both MODIS and HIRS/2-3 have ν_3 band coverage, but strong competing absorption by water vapor limits retrievals using these broadband sensors [*Prata et al.*, 2003]. The high spectral resolution of AIRS permits use of microwindows within the ν_3 band [e.g., *Mankin et al.*, 1992], providing high specificity to SO_2 (Figure 2) and better sensitivity than MODIS or HIRS/2-3 in the same region.

3. The October 2002 Eruption of Etna

[7] Etna's 2002 eruption, one of its more explosive in recent years, began late on 26 October, initially producing lava fountains and thick ash plumes from explosive vents (Global Volcanism Network (GVN), http://www.volcano.si.edu/volcanoes/region01/italy/etna/var_14.htm#bgvn_2711). Northerly winds lead to heavy ashfall south of Etna, forcing the closure of Catania airport on 27 October (GVN, http://www.volcano.si.edu/volcanoes/region01/italy/etna/var_14.htm#bgvn_2712). Fluxes of SO_2 measured by UV correlation spectrometry (COSPEC) averaged 10–16 Gg d^{-1} from late October to early December, with peak emissions of ~30 Gg d^{-1} in late November (GVN, http://www.volcano.si.edu/volcanoes/region01/italy/etna/var_14.htm#bgvn_2712), and ashfall was reported as far away as Libya and Greece (~350 and ~500 km from Etna respectively) [*Dellino and Kyriakopoulos*, 2003].

[8] Earth Probe TOMS retrievals of SO_2 abundance in the Etna eruption plumes were poor owing to the instrument's reduced sensitivity to lower tropospheric SO_2; GOME and SCIAMACHY retrievals are available at: http://www.oma.be/BIRA-IASB/Molecules/SO2/SO2_volc.php?cmd=init&volc=ETNA. At the time of writing, our AIRS retrievals are the only quantitative results for the 2002 Etna eruption obtained from a space-borne IR instrument of which we are aware.

4. Retrieval Procedures

[9] The Etna eruption plumes were located using bias difference (BD) images derived from AIRS channels on and off the spectral feature of interest (Figure 2). For volcanic clouds, modified versions of the radiative transfer algorithm SARTA (Standalone AIRS Radiative Transfer Algorithm) [*Strow et al.*, 2003] have been developed to retrieve SO_2 (using the ν_3 band) and ash column density, ash optical depth and particle size from AIRS spectra. SARTA is a stand-alone version of the AIRS RTA that allows fast radiative transfer calculations at the AIRS spectral resolution.

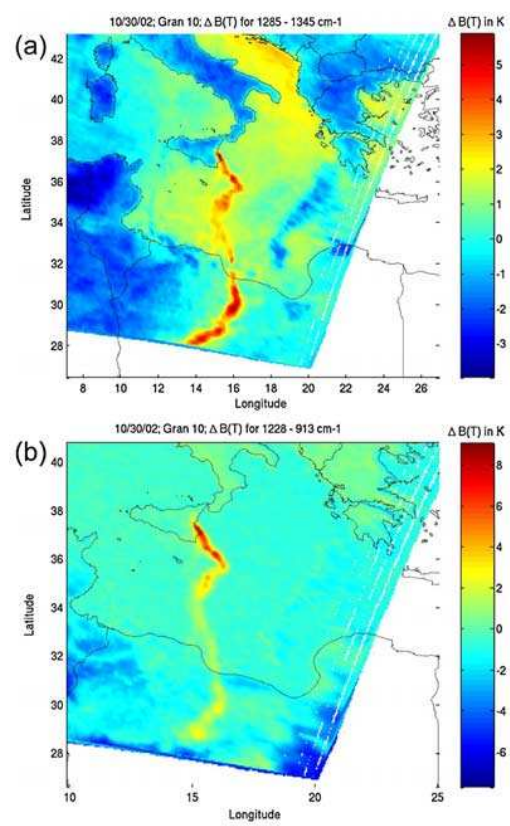

Figure 2. AIRS BD images of the Etna plume on 30 October 2002 at 0100 UT. Images are the difference in the bias between 2 channels; (a) 1285–1345 cm^{-1} bias showing SO_2 plume; (b) 1228–995 cm^{-1} bias showing aerosol plume. The bias is the difference between observed BTs and those computed from the ECMWF model fields for T(z) and H_2O(z).

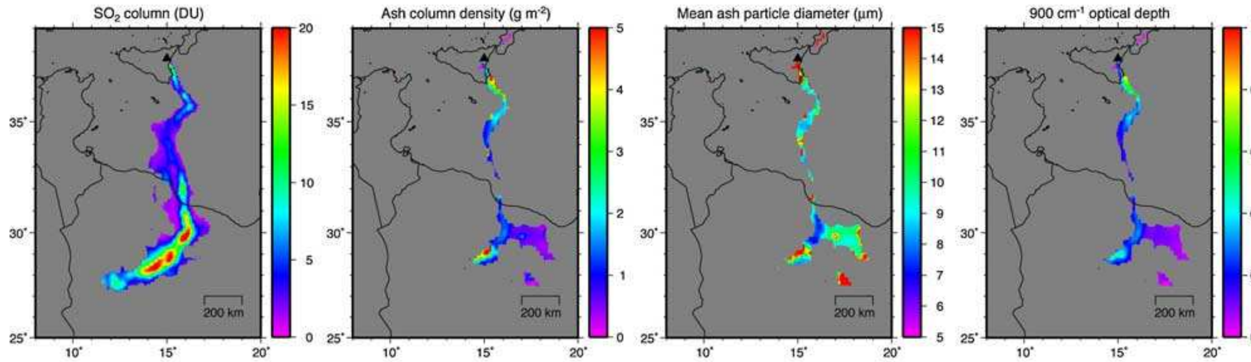

Figure 3. AIRS retrievals for the Etna plume on 30 October 2002 at 0100 UT. Total burdens at this time were ~24.5 Gg SO_2 and ~270 Gg ash (Table 1). Note the interference of low-level desert dust (indistinguishable from ash) in ash cloud retrievals over Libya.

[10] Vertical SO_2 column amounts were retrieved by placing a generic SO_2 profile at the nominal height of the volcanic plume, then iteratively running SARTA and scaling the profile until zero clear-sky bias was obtained in 5 AIRS channels across the ν_3 SO_2 band. The closest ECMWF analysis/forecast fields were used for the temperature and water profiles and low background BD values in Figure 2 indicate that the ECMWF profiles are quite accurate. We tentatively estimate the detection limit of the algorithm to be a total SO_2 column of ~3 Dobson Units (DU; 1 DU = 2.68×10^{16} molec cm^{-2}) in a single clear FOV, and retrieval error to be ~20% (excluding cloud height errors; see below). Further analysis of AIRS volcanic cloud data is required to develop a more rigorous error budget.

[11] A maximum cloud height of ~6.4 km was indicated by the Toulouse VAAC early in the Etna eruption, therefore the SO_2 perturbation was placed at 5–7 km for retrievals (a 2 km thick layer centered at 6 km). Note that the assumed height and temperature of the cloud exerts a strong influence on retrieved SO_2. Placing the cloud at an altitude of 7–9 km produced total SO_2 amounts 29–54% lower than those obtained for a cloud at 5–7 km. Retrievals using a SO_2 layer at 3–5 km altitude produced unreasonably high column amounts, and we conclude that the lower bound of AIRS vertical sensitivity lies within this range. A more sophisticated retrieval using the AIRS temperature and water vapor channels could provide more detailed information about SO_2 and ash vertical profiles, but this is beyond the scope of this paper.

[12] Ash retrievals assumed that the particles were of basaltic composition and spherical. Scattering parameters were calculated from Mie theory using a hybrid two-stream scattering algorithm and refractive indices (RIs) from *Pollack et al.* [1973]. A window of ~120 AIRS channels in the 700–1200 cm^{-1} range was used to retrieve ash column density, mean particle diameter and optical depth using SARTA with one scattering layer. We consider the mean particle diameter a rough estimate, especially for the larger particles that often occur in regions where the optical depth is small, which can easily lead to incorrect particle sizes.

[13] The assumed PSD influences ash retrievals. Both gamma and lognormal PSDs were evaluated, the latter obtained from an ash sample described in *Dellino and Kyriakopoulos* [2003]. Use of the gamma PSD typically produced a larger total ash mass, with an average positive bias of 32% relative to lognormal PSD retrievals. Particle size can vary by up to a factor of 2 according to the assumed PSD. Other parameters (RI, particle shape) will also impact retrievals but these quantities are poorly constrained for most ash types and the effects not well understood.

5. Results and Discussion

[14] Emissions from Etna were detected in AIRS BD images from 27 October–4 November (except on 31 October when winds were unfavorable), and were intermittently apparent until 12 November (subsequent AIRS data have not been examined in detail). The AIRS SO_2 signal began to wane on 1 November; coincident with a decline in activity (GVN, http://www.volcano.si.edu/volcanoes/region01/italy/etna/var_14.htm#bgvn_2712), and although elevated SO_2 emissions continued for ~1 month (GVN, http://www.volcano.si.edu/volcanoes/region01/italy/etna/var_14.htm#bgvn_2711) the plumes were evidently too low and/or too small (relative to the 13.5 km AIRS FOV) for detection. The presence of high cloud above the volcanic plume also impeded detection on several days.

[15] Results of retrievals for the period of vigorous activity from 27–30 October are shown in Figure 3, and in Table 1 with COSPEC SO_2 flux data from the Istituto Nazionale di Geofisica e Vulcanologia (INGV) for comparison. MODIS images on 28–30 October indicated high plume opacity close to the volcano due to dense ash and in these regions AIRS may underestimate burdens. The high sensitivity of AIRS permitted tracking of the plumes into N. Africa on most days, and the plume often remained remarkably homogeneous during transport despite a non-linear trajectory (e.g., Figure 2). Combining wind speeds measured during the eruption (~10–20 m s^{-1}) (S. Calvari, personal communication, 2003) with observed plume lengths (Table 1) suggests SO_2 lifetimes of ~1–2 days; consistent with prior loss rate determinations at Etna [e.g., *Martin et al.*, 1986] although the latter pertain to passive emissions. No major spatial separation was apparent between the SO_2 and ash streams, even in meandering plumes, although the ash plumes were typically shorter (e.g., Figure 2). We infer that the SO_2 and ash traveled at similar altitudes, with differing plume lengths reflecting depletion by discrete processes (i.e., oxidation of SO_2 vs. fallout of ash).

Table 1. SO_2 and Ash Mass Retrievals for Etna Eruption Plumes in October 2002

Date - Time (Local)[a]	AIRS SO_2 Mass (Mg)	COSPEC SO_2 Flux (Mg d^{-1})[b]	SO_2 Plume Extent (km)[c]	AIRS Ash Mass (Mg)[d]
10/27/02 – 1230-1236	15500[e]	11300	>430	23800[e]
10/28/02 – 0212-0218	24600		~1320	143200
10/28/02 – 1312-1318	47100	13300	~1420	92500
10/29/02 – 0118	29900[e]		>860	74200[e]
10/29/02 – 1354	15200[e]	20000	~1370	-
10/30/02 – 0200-0206	24500		~1340	266700
10/30/02 – 1300-1306	15400	19500	~1210	131700

[a]Time of AIRS data acquisition.
[b]Source: INGV eruption reports (http://www.ct.ingv.it/Etna2002/Geo&Vulca/Cospec/Cospec.htm). Note that timing of COSPEC data acquisition is not coincident with AIRS.
[c]Distance from Etna to the distal end of the plume detected by AIRS (measured along the plume); a minimum is given where the plume clearly continued beyond the edge of the AIRS data granule.
[d]Ash quantities given here assume a gamma PSD and a particle density of 2.75 g cm^{-3}.
[e]Denotes incomplete coverage of the plume, hence a minimum value.

[16] AIRS data indicate that, on occasions, concentrations of SO_2 and ash in the plumes did not decrease progressively downwind (e.g., Figure 3). Short-term variations in eruption discharge at the vent could explain this, although it could also reflect complex scavenging and removal mechanisms or release of SO_2 by re-evaporation of aerosol. These fluctuations complicate comparison of AIRS and COSPEC data (Table 1) since the latter cannot encapsulate short-term variability. Direct SO_2 column comparisons are also precluded as COSPEC traverses are made 7–15 km from Etna (Bruno et al., INGV reports, 2002, available at http://www.ct.ingv.it/Etna2002/Geo&Vulca/Cospec/Cospec.htm), where AIRS is unable to detect SO_2 due to low thermal contrast or insufficient plume altitude. Despite these limitations, AIRS total SO_2 is comparable to COSPEC SO_2 flux considering that some of the plumes may have contained more than one day's SO_2 load (Table 1). We also reiterate that errors in plume height assignment, and/or variations in plume height along its length, will affect retrieved SO_2 burdens. Scavenging of SO_2 by ash seems likely in view of the dense ash clouds emitted but will also have impacted the COSPEC data.

[17] Retrieval of ash properties in active eruption plumes has many applications, including aviation and health hazard mitigation, validation and initialization of trajectory models, and studies of ash fallout, aggregation and scavenging processes. AIRS retrievals indicate that the 2002 Etna plumes contained particles in the 10 μm size range (Figure 3), which is typical of fine volcanic ash and consistent with particle sizes measured in direct samples of Etna's eruptive emissions [*Allard*, 1999]. Particulate matter with diameter $\leq$10 μm (PM-10) is respirable and a known health hazard. The largest daily total ash mass measured by AIRS (~0.3 Tg; Table 1) is equivalent to ~10% of total PM-10 emissions from industrial and transportation sources in the U.S. in 1997 [*Environmental Protection Agency*, 1997].

[18] To demonstrate one potential application of AIRS data, we have derived ash fallout rates in the Etna plumes from the relationship between ash column density and plume age, the latter inferred from wind speeds (S. Calvari, personal communication, 2003). From a profile along a nocturnal plume observed at 0112UT on 28 October we estimate a mean ash fallout rate of ~1.7 g m^{-2} hr^{-1}. Profiles from daytime plumes give ash fallout rates of ~0.3–0.4 g m^{-2} hr^{-1} i.e., significantly lower than observed in the nocturnal plume. These rates are higher than predicted using a standard Stokes gravitational settling equation, which for a sphere with a diameter of 10 μm and a density of 2.5 g cm^{-3} yields a terminal velocity of ~0.01 m s^{-1}. Under these conditions ash particles would descend <0.5 km in 12 hours, but the ashfall recorded in Libya during the eruption suggests that particles descended several kilometers in <10 hours. We conclude that aggregation of ash particles was probably acting to increase ash removal rates. Faster removal of ash from the nocturnal plume may be promoted by increased condensation of water vapor onto ash particles and/or more widespread formation of ice in the plume during the night.

6. Conclusions

[19] AIRS will enhance estimates of tropospheric volcanic emissions during eruptions of moderate intensity, and its high spectral resolution could be used to derive vertical SO_2 profiles. The sensor is unlikely to detect most passive volcanic plumes owing to its relatively large FOV and the limitations of the ν_3 SO_2 band at low altitudes. Absorption bands of HCl and HF are not covered by AIRS, but CO_2 retrievals are possible (using absorption bands around 667 and 2326 cm^{-1}) and the potential for volcanic CO_2 detection is under investigation. Several issues require attention to improve ash retrievals, including better constraints on volcanic cloud PSDs and better treatment of non-spherical particles.

[20] **Acknowledgments.** We thank S. Calvari at INGV Catania for providing wind data. The comments of P. Allard and an anonymous referee significantly improved the paper.

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