

**Opportunistic
validation of sulfur
dioxide**

S. A. Carn and
T. M. Lopez

Opportunistic validation of sulfur dioxide in the Sarychev Peak volcanic eruption cloud

S. A. Carn¹ and T. M. Lopez²

¹Department of Geological and Mining Engineering and Sciences, Michigan Technological University, 1400 Townsend Drive, Houghton, MI 49931, USA

²Geophysical Institute, University of Alaska Fairbanks, 903 Koyukuk Drive, Fairbanks, AK 99775, USA

Received: 6 June 2011 – Accepted: 6 June 2011 – Published: 17 June 2011

Correspondence to: S. A. Carn (scarn@mtu.edu)

Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

We report attempted validation of Ozone Monitoring Instrument (OMI) sulfur dioxide (SO₂) retrievals in the stratospheric volcanic cloud from Sarychev Peak (Kurile Islands) in June 2009, through opportunistic deployment of a ground-based ultraviolet (UV) spectrometer (FLYSPEC) as the volcanic cloud drifted over Central Alaska. The volcanic cloud altitude (~12–14 km) was constrained using coincident CALIPSO lidar observations. By invoking some assumptions about the spatial distribution of SO₂, we derive averages of FLYSPEC vertical SO₂ columns for comparison with OMI SO₂ measurements. Despite limited data, we find minimum OMI-FLYSPEC differences of ~5–6 % which support the validity of the operational OMI SO₂ algorithm. These measurements represent the first attempt to validate SO₂ in a stratospheric volcanic cloud using a mobile ground-based instrument, and demonstrate the need for a network of rapidly deployable instruments for validation of space-based volcanic SO₂ measurements.

1 Introduction

Validation of satellite retrievals of trace gases is a crucial part of any mission, but the approach is highly dependent on the species in question. Some molecules (e.g., NO₂) have well-characterized sources and somewhat predictable distributions, making it easier to plan validation campaigns and extended deployments of ground-based or airborne instrumentation (e.g., Brinksma et al., 2008). However, this is not usually the case for volcanic SO₂, emitted by largely unpredictable volcanic activity, often in remote locations. Validation of satellite SO₂ measurements in such situations is mostly opportunistic (e.g., Carn et al., 2011), and may require rapid mobilization or deployment of ground-based assets (e.g., Spinei et al., 2010). Efforts to validate volcanic SO₂ retrievals have become increasingly important in the light of recent volcanic ash crises

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(e.g., the 2010 Eyjafjallajökull eruption), during which satellite measurements of SO₂ and ash were used to track the drifting volcanic clouds (e.g., Thomas and Prata, 2011).

We show here that useful SO₂ validation data can be collected by rapid deployment of a simple, mobile ultraviolet (UV) spectrometer system similar to those used widely for volcano monitoring. The opportunity arose when the volcanic cloud produced by the June 2009 explosive eruption of Sarychev Peak (Matua Island, Kuril Is; 48.1° N, 153.2° E) drifted over Central Alaska. Satellite measurements of SO₂ by the Ozone Monitoring Instrument (OMI) on NASA's Aura satellite were the object of validation. Our measurements represent the first attempt to validate SO₂ in a drifting, stratospheric volcanic cloud using road-based vehicular traverses beneath the plume.

The 2009 eruption of Sarychev Peak began on 11 June. Haywood et al. (2010) provide an overview of the eruption and present SO₂ data from the Infrared Atmospheric Sounding Interferometer (IASI) on the MetOp-A satellite, along with climate model simulations of volcanic cloud dispersion. IASI measured a total SO₂ burden of 1.2 ± 0.2 Tg in the upper troposphere and lower stratosphere (UTLS) following the eruption sequence, which is commensurate with total SO₂ burdens measured by OMI. Dispersion of the volcanic SO₂ after eruption was complex but is not the focus of this paper. An animation of OMI SO₂ data showing dispersion of the Sarychev Peak SO₂ cloud over the Northern Hemisphere from 10 June–31 July 2009 is available as Supplement. Beginning on 15 June, the SO₂ cloud began to drift across Alaska, providing the opportunity for validation described herein. In contrast to the SO₂ clouds released by the eruptions of Okmok and Kasatochi (Aleutian Islands) in July–August 2008 (e.g., Spinei et al., 2010), the bulk of the Sarychev eruption cloud resided at high latitudes, presenting few opportunities for ground-based measurements.

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2 Data

The Level 2 OMI dataset used here is derived from version 003 of the operational SO₂ algorithm (hereafter referred to as OMSO₂), the theoretical basis of which is described by Yang et al. (2007). We also use the Level 2 OMPIXCOR OMI Ground Pixel Corner Coordinate product. OMSO₂ and OMPIXCOR data are publicly available from the NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISC; <http://daac.gsfc.nasa.gov/>). Operational OMI SO₂ retrievals require an a-priori assumption of the vertical SO₂ distribution (Yang et al., 2007), which is currently addressed by providing retrieved SO₂ columns corresponding to four different a-priori SO₂ profiles. For this analysis we use the mid-tropospheric (TRM) and lower stratospheric (STL) SO₂ products, which correspond to SO₂ layer center of mass altitudes (CMAs) of ~7.5 and ~17.5 km, respectively. Overall uncertainty on the OMI SO₂ retrievals (including CMA errors) for SO₂ clouds above 5 km altitude is ~20% (Yang et al., 2007). We also employ other NASA A-Train satellite datasets, including Moderate Resolution Imaging Spectroradiometer (MODIS) data from Aqua, and Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) profiles from CALIPSO (Vaughan et al., 2004), to characterize meteorological clouds and assess volcanic cloud altitude (Figs. 1, 2).

Ground-based SO₂ data were collected using a FLYSPEC, a simple but flexible and rapidly deployable UV spectrometer that is calibrated using integral SO₂ gas cells (Horton et al., 2006). Conditions were favorable on 18 June 2009 as the Sarychev SO₂ cloud drifted over Southern and Central Alaska (Fig. 1; see Supplement). The FLYSPEC was mounted on a vehicle and pointed to zenith, and a high-quality instrument calibration was obtained under clear-sky conditions in Fairbanks (64.84° N, 147.72° W). Measurements of overhead SO₂ column density were then performed south of Fairbanks along Route 3 (Fig. 3). Conversion of calibrated FLYSPEC data (in ppm) to column densities in Dobson Units (DU; 1 DU = 0.02848 g m⁻² SO₂) used the conversion derived by Gerlach (2003) (1 ppm = 2.663 × 10⁻⁶ kg m⁻²). Further mobile and stationary FLYSPEC measurements were also made in Fairbanks on 20 June, when

AMTD

4, 3861–3875, 2011

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the Sarychev SO₂ cloud was again overhead. However, in this case the spatially extensive SO₂ cloud precluded FLYSPEC calibration under clear-sky conditions, and these measurements are not discussed further here. Noise in FLYSPEC SO₂ data has been reported to be ~0.4–1.1 DU, with errors of up to 6% on retrieved SO₂ columns based on testing with standard calibration cells (Elias et al., 2006).

3 Results

OMSO2 data show a filament of SO₂ extending across Central Alaska in the 22:28 UTC Aura overpass on 18 June 2009 (Fig. 1). CALIOP detected collocated aerosol (presumably sulfate) in the lower stratosphere at ~12–14 km altitude in the 22:22 UTC CALIPSO overpass (Fig. 2), while Aqua MODIS data indicate partly cloudy conditions and visible haze south of Fairbanks (Fig. 3). Figure 3 shows southbound and northbound FLYSPEC traverses superimposed on the OMI SO₂ retrievals. FLYSPEC traverse profiles, showing a relatively smooth, symmetrical plume cross-section in the southbound traverse, are shown in Fig. 3. The FLYSPEC traverses intersected five near-nadir OMI pixels in orbit 26207 (Fig. 3), and the average FLYSPEC SO₂ columns for each OMI pixel are reported in Table 1. We compute the OMI SO₂ column for SO₂ at 12–14 km altitude (we use 13 km) by linear interpolation between the OMSO2 TRM and STL columns. One factor that complicates the analysis is that the volcanic cloud was moving northwards during FLYSPEC data collection. Radiosonde soundings from Fairbanks (<http://weather.uwyo.edu/upperair/sounding.html>) at 12Z on 18 June and 00Z on 19 June indicate southerly to southeasterly winds at ~2.6–5.1 m s⁻¹ (9.4–18.5 km h⁻¹) at 13 km altitude. This is consistent with the observation that SO₂ was not detected over Fairbanks at ~21:10 UTC, but had drifted over Fairbanks by 00:15 UTC on 19 June and the northern flank of the plume was not detected in the northbound traverse (Figs. 3, 4). Although MODIS data indicate possible cloud interference, OMI cloud fractions were quite low in most pixels (with the exception of pixel P1; Table 1) and OMI cloud pressures imply predominantly low-altitude clouds which would not significantly

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impact OMI retrievals of stratospheric SO₂. The effect of clouds on the FLYSPEC measurements is unclear, but the general smoothness of the SO₂ cross-sections (Fig. 3a) suggests no significant cloud interference in most pixels.

Comparison between the OMI SO₂ columns and the average FLYSPEC SO₂ columns for each OMI pixel reveals some significant differences (Table 1), but the relative timing of the measurements and spatial averaging of SO₂ over the OMI pixels must be accounted for. In the absence of any ground-based constraints on horizontal variations in SO₂, we assessed the effects of spatial averaging by extending the FLYSPEC SO₂ columns laterally to simulate the 2-D distribution of SO₂ in the volcanic plume (Fig. 3d). The azimuth for this extrapolation is constrained using the location of maximum SO₂ in the FLYSPEC profile (64.27° N, 149.03° W; Fig. 4) and the location of the matching volcanic aerosol feature in CALIOP data (64.49° N, 145.96° W; Fig. 2), giving an azimuth of ~81°. This is qualitatively consistent with the SO₂ distribution mapped by OMI (Fig. 3). We then computed an unweighted mean of the simulated 2-D SO₂ columns over the OMI pixels (Table 1), resulting in adjustments of only a few percent relative to FLYSPEC traverse averages for some pixels, but more substantial (~10–130%; Table 1) for others.

Two important results arise from this analysis. Firstly, we find the best agreement between the OMI and spatially averaged FLYSPEC measurements for pixel P2 (13% difference; Table 1). We attribute this to the fact that pixel P2 has the best FLYSPEC data coverage of all the pixels (Fig. 3), providing the best constraints on the sub-pixel SO₂ distribution, despite a ~30–45 min time difference between the OMI and FLYSPEC measurements. The minimum time difference between OMI and FLYSPEC measurements (~5–15 min) occurred in pixel P5 (Fig. 3). However, we observe a ~90% difference between the OMI (3.1 DU) and FLYSPEC (6 DU) SO₂ columns on the southbound traverse, improving to ~40% for the northbound traverse when the average FLYSPEC SO₂ column (1.9 DU) is lower than OMI. Taking the mean of the southbound and northbound average FLYSPEC SO₂ columns (~4 DU) the difference between OMI and FLYSPEC SO₂ columns reduces to ~27%, which is close to the combined error

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on both datasets. We believe this approach is justified as neither the northbound nor the southbound FLYSPEC traverses correspond precisely in time to the OMI overpass, and the SO₂ cloud was drifting north at the time, thus shifting the region of higher SO₂ column amounts out of pixel P5 and leading to the reduction in FLYSPEC SO₂ column.

Differences between OMI and FLYSPEC SO₂ columns are larger for the other analyzed pixels, particularly around the location of maximum SO₂ (Table 1; Fig. 4a). This is perhaps best explained by spatial heterogeneity in the volcanic cloud that is not captured in our simple 2-D model of the plume (Fig. 3d). Furthermore, pixels P3 and P4 were not well-covered by the FLYSPEC data and the traverses were close to the pixel boundaries, where the precise definition of the OMI spatial resolution becomes critical due to the variable OMI pixel spatial response function (SRF). All these factors may have contributed to the lower SO₂ column measured by OMI in these pixels. It is likely that the larger cloud fraction in pixel P1 caused the relatively poor agreement for the southbound traverse (Fig. 3; Table 1). Increasing discrepancies are seen for the northbound FLYSPEC traverse data (Fig. 4b) as by this time the SO₂ cloud had moved north over Fairbanks.

It is important to note here that the spatial resolution of OMI (or any CCD-array based spectrometer) cannot be simply represented by the mapped pixel edges (Figs. 3, 4). The SRF in both the flight- and across-track direction is approximately Gaussian in shape (Dobber et al., 2006). More precisely, the spatial resolution in the flight direction (~ 13 km at nadir) is defined as the full-width at half-maximum (FWHM) of the telescope instantaneous field-of-view (IFOV; $\sim 1^\circ$) convolved with a 2-s integration time, while in the across-track direction it is the FWHM of the sum of the Gaussian SRFs of 8 binned OMI CCD pixels (~ 24 km at nadir). Therefore, each OMI pixel is also influenced by photons received from beyond the mapped pixel boundaries. A detailed analysis of the OMI spatial resolution is beyond the scope of this paper, but we assessed the effect of non-uniform spatial response by weighting the FLYSPEC SO₂ columns with a flat-topped Gaussian function (of the form $f(x) = \exp(-c(x - x_0)^4)$, where $x - x_0$ is absolute distance from the pixel center (x_0) in the along-track direction, and c is a constant) to

simulate an along-track SRF with FWHM corresponding to the mapped pixel boundaries. No attempt was made to account for contributions from beyond the FWHM pixel boundary, or to simulate the across-track SRF. The resulting weighted, spatially averaged FLYSPEC SO₂ columns are ~ 13–24% lower than the unweighted averages (Table 1) and improve the comparisons. For pixel P2, the OMI-FLYSPEC difference reduces to ~ 6%, whilst for pixel P5, the OMI-FLYSPEC difference for the mean of the northbound and southbound traverses reduces to ~ 5%, both well within measurement error. Significant differences remain for the other pixels, which we conclude is probably due to unconstrained sub-pixel spatial heterogeneity in the volcanic SO₂ cloud.

4 Conclusions

As this study demonstrates, validation of volcanic SO₂ measurements can be challenging, since acquisition of high-quality ground-based data may be precluded by time constraints. Nevertheless, we find good agreement between selected OMI and spatially averaged FLYSPEC SO₂ data when some necessary assumptions about the 2-D SO₂ distribution are invoked, providing additional support for the validity of the operational OMSO2 dataset (see also Spinei et al., 2010; Carn et al., 2011). Unresolved spatial heterogeneity in the volcanic cloud on a sub-OMI pixel scale appears to be the best explanation for other observed OMI-FLYSPEC differences.

We conclude by stressing the uniqueness of our FLYSPEC SO₂ dataset for the Sarychev Peak eruption, one of the last decade's largest explosive eruptions. As discussed above, the Sarychev volcanic cloud mostly resided at high Arctic latitudes, precluding widespread ground-based measurements. Perro et al. (2010) report ground-based Brewer spectrophotometer detection of SO₂ from Sarychev over Eureka, Northern Canada on 1 July 2009. To our knowledge, the FLYSPEC SO₂ data presented here are the closest measurements to the eruption in time and location. The successful ground-based measurement of the volcanic SO₂ cloud underlines the need for

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a widespread network of rapidly deployable instruments in order to successfully validate volcanic SO₂ retrievals from satellite sensors.

Supplementary material related to this article is available online at:

<http://www.atmos-meas-tech-discuss.net/4/3861/2011/>

5 **[amtd-4-3861-2011-supplement.zip](#)**.

Acknowledgement. Funding for this work was provided by NASA (award no. NNX09AJ40G for Aura validation).

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Table 1. OMSO2 (orbit 26207; 22:28 UTC) and FLYSPEC data for Sarychev plume pixels on 18 June 2009.

Pixel No.	xT ¹	Lat (° N) ²	Lon (° W) ²	OMI SO ₂ column (DU)			Average FLYSPEC SO ₂ column (DU)		Cloud fraction ⁸
				TRM ³	STL ⁴	Interp. ⁵	S. Traverse ⁶	N. Traverse ⁷	
P1	29	64.70	148.47	3.6	3.4	3.5	1.0 (2.4, 2.1)	14.2 (14.0, 12.3)	0.57
P2	28	64.63	148.93	4.2	3.8	4.0	5.7 (4.5, 3.7)	13.8 (13.9, 10.6)	0.34
P3	28	64.51	148.83	7.8	6.9	7.3	14.5 (14.7, 12.6)	13.9 (12.5, 10.5)	0.25
P4	27	64.44	149.29	7.6	6.7	7.1	15.3 (14.5, 12.4)	10.0 (9.8, 8.3)	0.25
P5	27	64.32	149.18	3.3	3.0	3.1	6.3 (6.0, 5.0)	2.0 (1.9, 1.6)	0.27

¹ OMI cross-track pixel position (1–60; 30 is nadir).

² Lat, Lon corresponds to center of OMI pixel.

³ Collection 3 OMSO2 data for a SO₂ CMA of 7.5 km (TRM product).

⁴ Collection 3 OMSO2 data for a SO₂ CMA of 17.5 km (STL product).

⁵ TRM and STL OMSO2 columns linearly interpolated to a SO₂ cloud altitude of 13 km.

⁶ Average FLYSPEC SO₂ column for southbound traverse from Fairbanks. Data in parentheses account for unweighted and weighted spatial averaging across the OMI pixel, respectively.

⁷ Average FLYSPEC SO₂ column for northbound traverse towards Fairbanks. Data in parentheses account for unweighted and weighted spatial averaging across the OMI pixel, respectively.

⁸ OMI-derived cloud fraction in pixel.

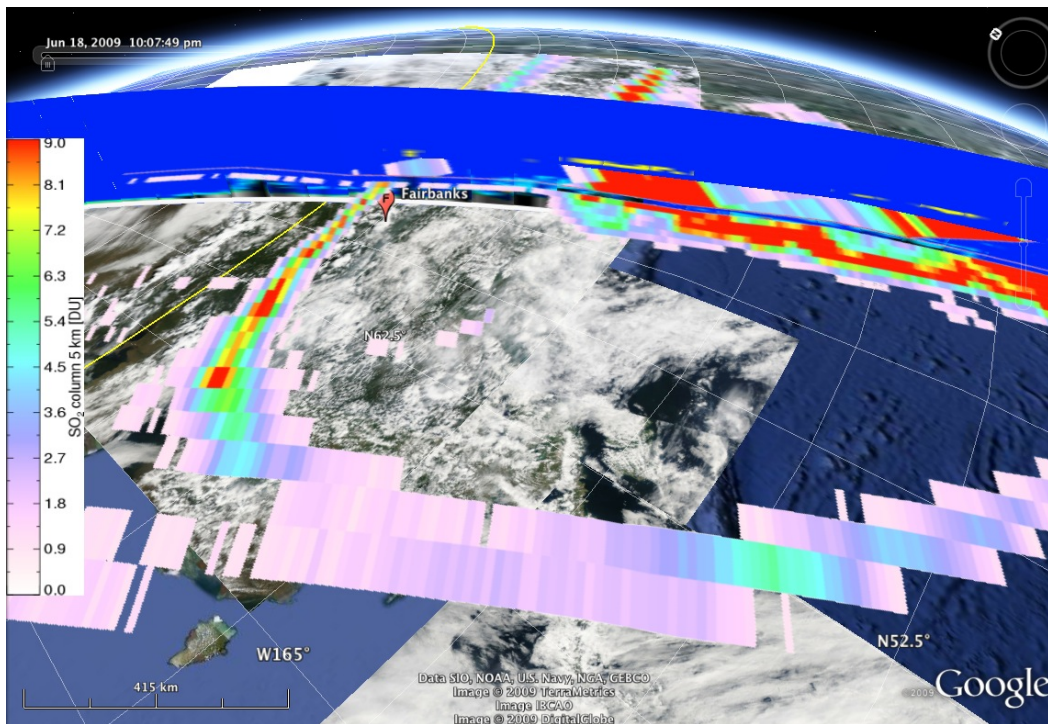


Fig. 1. Google Earth™ composite showing a perspective view of A-Train satellite observations of the Sarychev Peak volcanic cloud over Alaska on 18 June 2009. The datasets shown are true-color Aqua MODIS visible imagery (~ 22:20 UTC; lowermost image), OMI SO₂ columns (~ 22:28 UTC; large colored pixels – color scale shown on left) and a CALIPSO lidar Vertical Feature Mask (VFM) curtain (~ 22:20 UTC). Yellow features on the CALIPSO VFM curtain indicate stratospheric volcanic aerosol at altitudes of ~ 12–15 km. The location of Fairbanks, AK is indicated.

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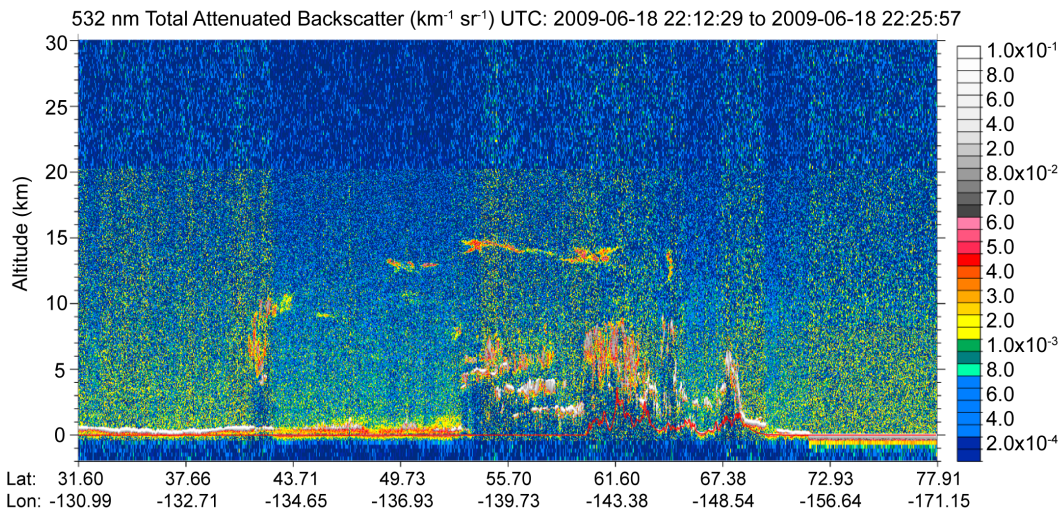


Fig. 2. 532 nm total attenuated backscatter curtain from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) aboard the CALIPSO satellite, collected at $\sim 22:12\text{--}22:26$ UTC on 18 June 2009. This plot and other coincident CALIOP data can be seen at: http://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_date.php?s=production&v=V3-01&browse_date=2009-06-18. Features showing elevated backscatter at altitudes above ~ 11 km are probably all due to aerosol in the Sarychev Peak volcanic cloud, based on collocation with SO_2 measured by OMI (Fig. 1). Low values of the lidar depolarization ratio in these features (not shown) suggest that the aerosol was dominated by liquid sulfate aerosol and/or liquid-coated solid particles. The volcanic cloud filament discussed in this paper is the northernmost volcanic aerosol feature at $\sim 64\text{--}65^\circ$ N.

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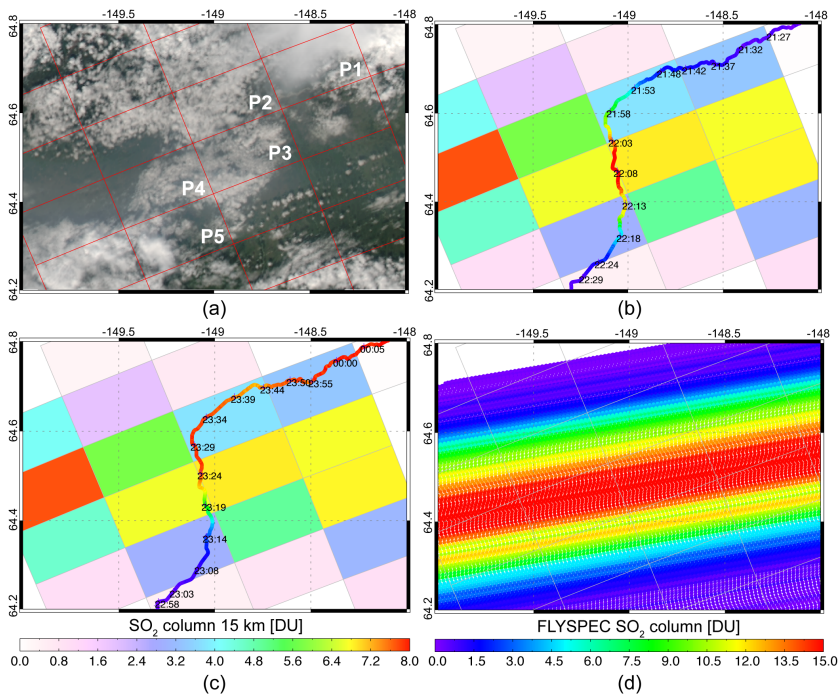


Fig. 3. (a) Aqua MODIS visible image of the region south of Fairbanks, AK on 18 June 2009 at 22:20 UTC with OMI pixel boundaries overlain in red (pixel dimensions are $\sim 13 \times 24$ km). OMI pixel numbers correspond to those in Table 1. Note visible haze in the volcanic cloud across image center. (b) OMI SO_2 columns (22:28 UTC; colored pixels) and ground-based FLYSPEC SO_2 traverse southbound from Fairbanks (periodically time stamped in UTC). OMI SO_2 color bar is shown in (c); FLYSPEC data color bar is shown in (d); (c) OMI SO_2 columns (22:28 UTC) and ground-based FLYSPEC SO_2 traverse northbound towards Fairbanks; (d) Lateral extrapolation of southbound traverse FLYSPEC data to assess effects of spatial averaging. OMI pixel boundaries are overlain in gray.

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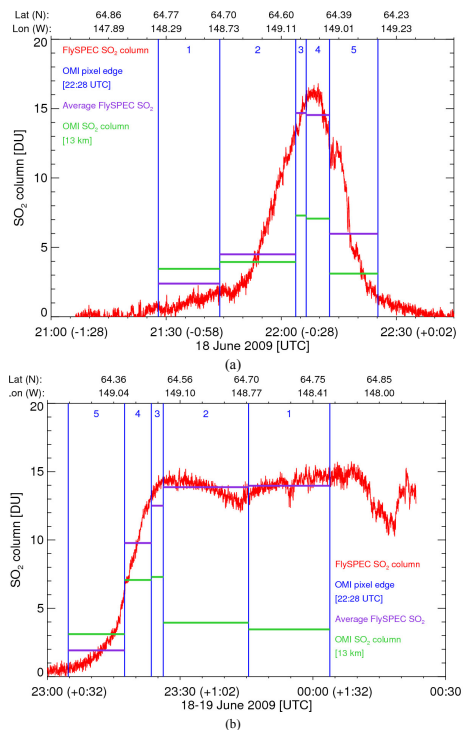


Fig. 4. FLYSPEC SO₂ columns measured during traverses south of Fairbanks (64.84° N, 147.72° W) along Route 3 (red line), OMI pixel boundaries for the 22:28 UTC Aura overpass (vertical blue lines; pixel numbers referred to in Table 1 are shown at top), FLYSPEC SO₂ columns averaged (unweighted) over corresponding OMI pixels (horizontal purple lines) and interpolated OMI SO₂ columns in the same pixels (horizontal green lines). The abscissa shows FLYSPEC time; time relative to the Aura overpass is indicated in parentheses. **(a)** Southbound traverse from Fairbanks; **(b)** northbound traverse towards Fairbanks. The absence of a peak in **(b)** is due to northward drift of the volcanic cloud during measurement.

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