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► **To cite this version:**

S. Martínez-Alonso, M. N. Deeter, H. M. Worden, Cathy Clerbaux, D. Mao, et al.. First satellite identification of volcanic carbon monoxide. *Geophysical Research Letters*, 2012, 39 (21), pp.L21809. 10.1029/2012GL053275 . hal-00750411

**HAL Id: hal-00750411**

**<https://hal.science/hal-00750411>**

Submitted on 16 Mar 2016

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## First satellite identification of volcanic carbon monoxide

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Received 26 July 2012; revised 28 September 2012; accepted 2 October 2012; published 9 November 2012.

[1] Volcanic degassing produces abundant H<sub>2</sub>O and CO<sub>2</sub>, as well as SO<sub>2</sub>, HCl, H<sub>2</sub>S, S<sub>2</sub>, H<sub>2</sub>, HF, CO, and SiF<sub>4</sub>. Volcanic SO<sub>2</sub>, HCl, and H<sub>2</sub>S have been detected from satellites in the past; the remaining species are analyzed *in situ* or using airborne instruments, with all the consequent limitations in safety and sampling, and at elevated costs. We report identification of high CO concentrations consistent with a volcanic origin (the 2010 Eyjafjallajökull and 2011 Grímsvötn eruptions in Iceland) in data from the Measurements of Pollution in the Troposphere instrument (MOPITT) onboard EOS/Terra. The high CO values coincide spatially and temporally with ash plumes emanating from the eruptive centers, with elevated SO<sub>2</sub> and aerosol optical thickness, as well as with high CO values in data from the Infrared Atmospheric Sounding Interferometer (IASI), onboard MetOp-A. CO has a positive indirect radiative forcing; climate models currently do not account for volcanic CO emissions. Given global volcanic CO<sub>2</sub> emissions between 130 and 440 Tg/year and volcanic CO:CO<sub>2</sub> ratios from the literature, we estimate that average global volcanic CO emissions may be on the order of ~5.5 Tg/year, equivalent to the CO emissions caused by combined fossil fuel and biofuel combustion in Australia. **Citation:** Martínez-Alonso, S., M. N. Deeter, H. M. Worden, C. Clerbaux, D. Mao, and J. C. Gille (2012), First satellite identification of volcanic carbon monoxide, *Geophys. Res. Lett.*, 39, L21809, doi:10.1029/2012GL053275.

### 1. Introduction

[2] Volcanic gas emissions before, during, and after terrestrial eruptions commonly include H<sub>2</sub>O, CO<sub>2</sub>, SO<sub>2</sub>, HCl, H<sub>2</sub>S, S<sub>2</sub>, H<sub>2</sub>, HF, CO, and SiF<sub>4</sub> [Symonds *et al.*, 1994]; the first two constitute the largest part of volcanic emissions. The relative composition and rate of release of these volatiles contain keys to understanding the eruptive style and predicting volcanic events [Symonds *et al.*, 1994; Thomas and Watson, 2010]. Some volcanic gases (H<sub>2</sub>O, CO<sub>2</sub>, CO) have a direct or indirect positive radiative forcing and thus impact climate [Forster *et al.*, 2007].

[3] Volcanic gases were traditionally sampled *in situ* and, subsequently, in airborne campaigns. The latter are costly; both are spatially and temporally constricted as well as

hazardous. Satellite detection of volcanic gases, which would be mostly free from these drawbacks, has been achieved for only a few species. Detection of volcanic water vapor and CO<sub>2</sub> is challenging due to high background levels of these gases in the atmosphere [Symonds *et al.*, 1994]. In contrast, due to its relative high abundance in volcanic plumes and very low background levels, volcanic SO<sub>2</sub> is routinely analyzed from satellite data [Oppenheimer *et al.*, 2011, and references therein]. Other volcanic species such as HCl [Prata *et al.*, 2007] and H<sub>2</sub>S [Clarisse *et al.*, 2011] have also been successfully identified from satellites.

[4] To test the feasibility of volcanic CO detection from satellite we have focused our efforts on the analysis of several datasets acquired over the Iceland region (58°N, -28°E to 68°N, -10°E) during the Eyjafjallajökull 2010 and Grímsvötn 2011 eruptions.

### 2. Observations

[5] In this study we use satellite observations acquired by the MOPITT, MODIS, IASI, and OMI instruments. MOPITT is a nadir-looking, cross-track scanning infrared radiometer onboard EOS/Terra that uses gas correlation spectroscopy to detect CO in the troposphere. We used version 4, level 2 MOPITT CO retrievals obtained from its thermal infrared (TIR) channels (7D, 5D, and 5A; D and A channels are sensitive to target-gas and background information, respectively). These channels sense incoming radiation in the 4.56 to 4.67  $\mu\text{m}$  spectral range, coinciding with the R-branch of the CO rotational-vibrational fundamental mode. MOPITT provides total CO column values as well as some information on CO vertical distribution. Global MOPITT coverage is achieved in approximately 3 days; its ground instantaneous field of view (GIFOV) is 22 by 22 km<sup>2</sup> at nadir. We used day-only, cloud-free, otherwise unfiltered MOPITT data. To maximize the number of MOPITT retrievals, cloud detection was based on MOPITT TIR radiances only; this did not distort the extent of the plumes or their retrieved CO values. MOPITT data were corrected for a systematic geolocation error by shifting the reported longitude 0.35° to the east (web3.acd.ucar.edu/mopitt/GeolocationBiasReport.pdf).

[6] To locate volcanic plumes and contrast them with MOPITT CO values we inspected simultaneously acquired EOS/Terra MODIS (Moderate Resolution Imaging Spectroradiometer) true color images with an effective GIFOV of 0.5 by 0.5 km<sup>2</sup> at nadir. EOS/Terra MODIS aerosol optical thickness (AOT) at 0.55  $\mu\text{m}$  was also analyzed; level 2 products with a 10 by 10 km<sup>2</sup> GIFOV from version 051 processed after 28 September 2010 (thus not affected by an error involving incorrect clear sky radiances) were utilized.

[7] IASI total CO column data (level 2, version v20100815) for dates with anomalous MOPITT CO values and visible

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volcanic plumes were also analyzed. MOPITT and IASI apply different measurement techniques and retrieval algorithms to derive CO abundances, thereby offering two independent sets of evidence to investigate the detectability of volcanic CO from satellite. IASI is a nadir looking Fourier transform infrared spectrometer onboard MetOp-A. It covers the 3.6–15.5  $\mu\text{m}$  spectral range sampling every 0.25  $\text{cm}^{-1}$ , and thus can resolve individual absorption lines of CO and other atmospheric infrared active species. Global coverage is achieved twice daily with a 12 km diameter GIFOV at nadir. IASI retrievals are commonly filtered for clouds based on Eumetsat cloud information; additionally, we used coeval MetOp-A AVHRR/3 data to ensure that only cloud-free IASI pixels were utilized in this analysis. We also analyzed IASI brightness temperature (BT) spectra in the MOPITT TIR channel region to derive the depth of diagnostic CO lines, which is related to CO abundance, and to investigate the radiative effects of volcanic aerosols.

[8]  $\text{SO}_2$  is a well established atmospheric volcanic marker. We analyzed total  $\text{SO}_2$  column values (data set version 003, level 2) obtained from measurements by the Ozone Monitoring Instrument (OMI) onboard EOS/Aura. OMI provides daily global coverage with an GIFOV of 24 by 13  $\text{km}^2$  at nadir. For each OMI scene lower troposphere (TRL) and mid troposphere (TRM) operational  $\text{SO}_2$  column density values are provided; these values are obtained assuming a center of mass altitude (CMA) for the volcanic plume at 2.5 and 7.5 km, respectively. Column density values for CMA between these can be obtained by linear interpolation ([http://so2.gsfc.nasa.gov/Documentation/OMSO2Readme\\_V111\\_0818.htm](http://so2.gsfc.nasa.gov/Documentation/OMSO2Readme_V111_0818.htm)). We discarded pixels with radiative cloud fraction  $\geq 0.2$ .

[9] The altitude of the volcanic cloud during the 2010 eruption was constrained using measurements from the C-band weather radar located in Keflavík International Airport [Arason *et al.*, 2011]. A report describing the 22 May 2011 eruption contains plume height estimates based on similar measurements [Jakobsdóttir *et al.*, 2012].

### 3. Results

[10] We have identified clusters of anomalously high MOPITT CO values on 19 April 2010 and 7 May 2010 (Eyjafjallajökull eruption), as well as on 22 May 2011 (Grímsvötn eruption). We consider anomalous values the highest retrievals (on a specific date, in the study area) which coincide spatially with a volcanic plume in simultaneously acquired MODIS visible data. Similarly, we consider background values the retrievals outside that plume. Anomalous and background values represent two separate populations, according to their value and spatial distribution.

[11] The anomalous MOPITT CO values on the three dates are spatially continuous and have a plume-like distribution (Figure 1, left). The reported average height for the top of the volcanic plume during satellite overpass was 2.5 km (19 April) and 5.3 km (7 May) [Arason *et al.*, 2011]. On 22 May the lower boundary of the volcanic plume to the south of the vent was at about 5 km [Jakobsdóttir *et al.*, 2012]. Compelling evidence of further anomalous values during the remaining dates of volcanic activity in 2010 and 2011 was not found due to either meteorological clouds precluding the CO retrievals or to lack of sufficient MOPITT coverage. We calculate the difference between MOPITT

total CO column in the plume versus that in the background to be between 4.48 and  $4.05\text{e} + 017 \text{ mol/cm}^2$  in the Eyjafjallajökull eruption and  $1.89\text{e} + 017 \text{ mol/cm}^2$  in the Grímsvötn eruption (Table S1 in the auxiliary material).<sup>1</sup>

[12] IASI CO total column data on 7 May 2010 show a plume-like set of anomalously high values which coincide spatially with these shown by MOPITT. Their mean exceeds the average CO background value by  $6.38\text{e} + 017 \text{ mol/cm}^2$  (Figure 2 and Table S1). IASI data on 19 April 2010 and 22 May 2011 over the plumes were deemed cloudy according to the Eumetsat cloud information (i.e., cloud coverage in the pixel  $\geq 12\%$ ), and thus sufficient spatial coverage for these two dates is not available.

[13] On the three dates analyzed the anomalous MOPITT CO values show good overlap with volcanic plumes apparent in the MODIS visible images and with high AOT values (Figure 1, middle). MODIS measured a wide range of AOT values over the MOPITT CO plumes: 0.2 to 2.96 (19 April 2010), 0.09 to 1.54 (7 May 2010), and 0.001 to 2.7 (22 May 2011).

[14] OMI shows  $\text{SO}_2$  plumes emanating from the volcanic centers on all three dates (Figure 1, right). We have derived  $\text{SO}_2$  column values by linear interpolation between operational OMI TRL and TRM retrievals, for a CMA equal to the reported height of the top (or bottom, depending on availability) of the volcanic cloud during satellite overpass. By using the height of the cloud's top rather than the center of mass altitude the  $\text{SO}_2$  column values are slightly underestimated ( $\sim 8.5\%$ , for a 2 km thick cloud). Using the clouds' bottom results in a similar overestimation. We calculate an average  $\text{SO}_2$  total column value of  $5.76\text{e} + 017$  (plume directly south of the vent on 19 April 2010),  $1.12\text{e} + 018$  (7 May 2010), and  $3.75\text{e} + 018 \text{ mol/cm}^2$  (22 May 2011), or 2.14, 4.17, and 13.96 DU (Dobson Units), respectively. These measurements are well above OMI's detection limit [OMI Team, 2012].

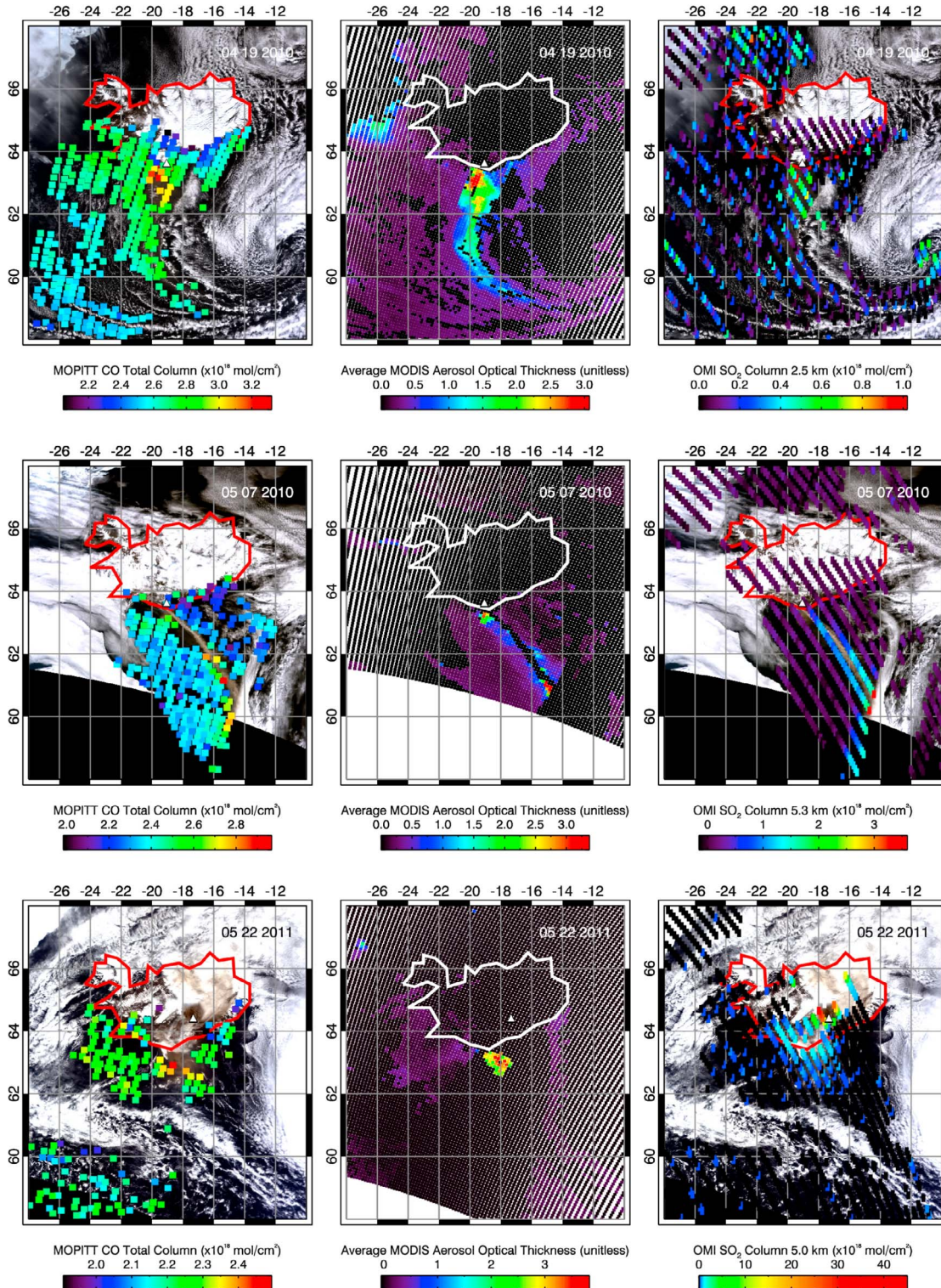
## 4. Discussion

### 4.1. Artifact or Volcanic CO?

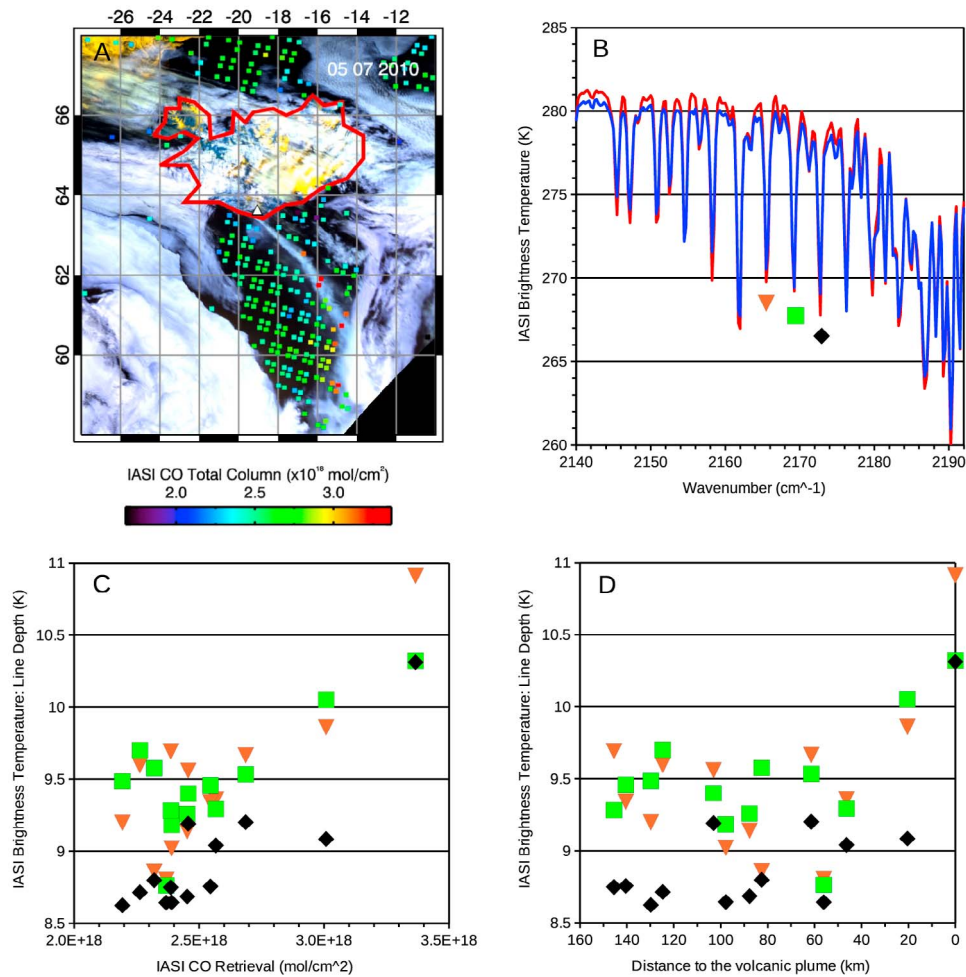
[15] In order to determine if the anomalous MOPITT CO values in the volcanic plumes is an artifact produced by other volcanic gases and/or volcanic aerosols we have investigated other sources of absorption in the MOPITT TIR bandpass. Due to the nature of gas correlation radiometry, non-target gases have very little effect on the measured radiances. Additionally, most relevant volcanic gases have no strong absorption lines in the MOPITT TIR bandpass (e.g.,  $\text{CO}_2$ ,  $\text{SO}_2$ , HCl,  $\text{H}_2\text{S}$ ,  $\text{S}_2$ ,  $\text{H}_2$ , HF,  $\text{SiF}_4$ , SO,  $\text{N}_2$ ,  $\text{CH}_4$ , Ar,  $\text{O}_2$ , and  $\text{NH}_3$ ). The opposite is true for water vapor and carbonyl sulfide (a minor volcanic gas); however, the latter is nearly two orders of magnitude less abundant than CO in average (from data in Symonds *et al.* [1994]) and thus would produce a much weaker signal.

[16] To model the sensitivity of MOPITT CO retrievals to elevated water vapor we used MOPABS (the MOPITT absorption model [Edwards *et al.*, 1999]). We ran simulations with a standard atmosphere input and then modified that input to include: 1) a CO profile increased by 10% at all pressure levels, 2) the average background NCEP/GDAS

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2012GL053275.



**Figure 1.** Volcanic markers mapped for the three dates analyzed. Iceland’s contour and the location of the active volcanic vent (white triangle) are shown for clarity. The size of the colored pixels is that of the instrument’s GIFOV, unless noted otherwise. (left) MOPITT total CO column values over coeval MODIS true color image. Anomalously high CO values over the volcanic plume appear in yellow and red. (middle) MODIS AOT. Plume values appear in cyan, green, yellow, and red. (right) OMI total SO<sub>2</sub> column over coeval MODIS true color image. Anomalously high SO<sub>2</sub> values appear in cyan, green, yellow, and red. OMI pixels shown enlarged (×2) for clarity.



**Figure 2.** (a) IASI total CO column values on 7 May 2010 over coeval AVHRR/3 image. Anomalous high CO values in the volcanic plume appear in orange and red. IASI scanlines run west-northwest to east-southeast. (b) IASI BT spectra acquired in one scanline inside (red) and away from (blue) the volcanic plume. The spectral range shown is that of the MOPITT TIR channels; selected CO absorption lines (at 2165.5, 2169.2, and 2172.7 cm<sup>-1</sup>) are indicated with colored symbols. Slight BT differences between spectra continuum (less than 1 K in the example shown) are not systematic and could be due to meteorological effects, among others. (c) Depth of CO lines in IASI BT spectra acquired in one scanline versus IASI CO retrievals (symbols as in Figure 2b). (d) Depth of CO lines in IASI BT spectra acquired in one scanline versus distance to the volcanic plume (symbols as in Figures 2b).

(National Centers for Environmental Prediction/Global Data Assimilation System) water vapor profile for 7 May 2010, and 3) the average NCEP water vapor profile for the plume locus on that same date. (NCEP/GDAS water vapor profiles, which are utilized in operational MOPITT CO retrievals, are derived from multi-sensor measurements; climatological

data are utilized to complement lacking or insufficient measurements. We have corroborated that water vapor profiles in the volcanic plume and its surroundings do not include climatological data.) Modeling results (Table 1) show that the change in MOPITT TIR D signal due to increased water vapor in the plume is 1 to 2 orders of

**Table 1.** MOPABS Sensitivity Test Results: Effect on MOPITT TIR Radiances of a 10% CO Increase (the MOPITT Detection Limit) Versus That Produced by Differences in Water Vapor Between the Volcanic Plume and Its Surroundings<sup>a</sup>

|                                     | MOPABS Radiance (W m <sup>-2</sup> sr <sup>-1</sup> ) |                       |                       |
|-------------------------------------|---|-----------------------|-----------------------|
|                                     | MOPITT TIR Channel 7D                                 | MOPITT TIR Channel 5D | MOPITT TIR Channel 5A |
| Standard atmosphere                 | 2.11064E-4  | 1.31012E-2            | 4.55677E-2            |
| 10% CO enriched atmosphere          | 2.02425E-4  | 1.30718E-2            | 4.55336E-2            |
| Water vapor profile: background     | 2.11615E-4  | 1.31502E-2            | 4.57532E-2            |
| Water vapor profile: plume          | 2.11579E-4  | 1.31450E-2            | 4.57294E-2            |
| D/A change: standard vs CO enriched | 4.09%   | 0.22%                 | -                     |
| D/A change: background vs plume     | 0.02%   | 0.04%                 | -                     |

<sup>a</sup>Water vapor profiles are NCEP averages for 7 May 2010.

magnitude below that due to a 10% change in CO, which is the MOPITT detection limit [Pan *et al.*, 1995].

[17] Volcanic aerosols are strong absorbers at longer wavelengths (i.e., near 10  $\mu\text{m}$ ) [Clarisse *et al.*, 2010]; their behavior in the MOPITT TIR bandpass is less well understood. To investigate this point we analyzed IASI BT spectra on 7 May 2010, focusing on sets of cloud-free, adjacent spectra acquired in the same scanline inside of and away from the volcanic plume. Our analysis shows that in the spectral region of the MOPITT TIR bandpass, aerosol-free and aerosol-rich spectra (from background and plume, respectively) are indistinguishable in terms of the shape of their continuum and individual absorption lines (Figure 2b). Three of the diagnostic CO absorption lines in this spectral range (at 2165.5, 2169.2, and 2172.7  $\text{cm}^{-1}$ ) were selected for further analysis, based on their intensity and lack of overlap with H<sub>2</sub>O lines. Their depth was quantified and contrasted to IASI CO values and distance to the volcanic plume. As expected, line depth increases with CO abundance (Figure 2c). For distances less than 40 km, line depth and thus CO abundance are inversely proportional to distance between the spectrum locus and the plume (Figure 2d). For distances greater than this threshold line depth and distance show no correlation. This behaviour of the CO spectral features with respect to proximity of the plume is consistent with a volcanic origin for the anomalously high CO values in the plume.

#### 4.2. Satellite Detection of Volcanic CO Versus Aircraft Observations and Other Volcanic Markers

[18] The maximum MOPITT and IASI CO values in the plume differ in magnitude by less than 14% ( $2.97\text{e} + 018$  and  $3.37\text{e} + 018$   $\text{mol}/\text{cm}^2$ , respectively, on 7 May 2010). MOPITT mixing ratio profile values are also close in magnitude to airborne CO measurements during the 2010 eruption (CARIBIC project [Brenninkmeijer *et al.*, 2007] and DLR-Falcon campaign [Schumann *et al.*, 2011]). CARIBIC flights on 20 April, 16 May, and 19 May 2010 intersected the volcanic plume at an approximate altitude of 4 km (600 hPa) over northern Germany, Northern Ireland and the Norwegian Sea, respectively. The average CO background values were near 120 ppbv, while values in the plume for each of the three dates reached 200, 205, and 179 ppbv respectively [Rauthe-Schöch *et al.*, 2012]. The flights in the DLR-Falcon campaign sampled a fresh plume between Iceland and northern Scotland on 2 May at 3.5 km altitude (661 hPa), with background and plume CO values of 128.5 and 200.9 ppbv, respectively. Despite the spatial and temporal differences, we find that in most cases airborne CO measurements and MOPITT mixing ratio values derived for similar pressure levels (Table S1) agree within 10%.

[19] As shown by OMI measurements, volcanic SO<sub>2</sub> plumes are present on all three dates analyzed and coincide spatially with the MOPITT and IASI CO plumes, as well as with high MODIS AOT values. On 19 April 2010 a second cluster of anomalously high SO<sub>2</sub> values was detected away from the vent, near 59°N, -11°E; this would be consistent with an older volcanic cloud transported SE by the prevalent winds. We find that average SO<sub>2</sub> total column in the plume is roughly of the same magnitude (19 April 2010) or one order of magnitude above (7 May 2010 and 22 May 2011) the average MOPITT CO total column values. This is consistent with CO:SO<sub>2</sub> ratios obtained from field and airborne

measurements in volcanic environments where tectonic plates diverge, such as Iceland [Symonds *et al.*, 1994; Oppenheimer *et al.*, 2011].

#### 5. Summary and Conclusions

[20] We report satellite identification of volcanic CO for the first time, achieved using data from two independent instruments: MOPITT and IASI. Other volcanic markers (ash plume in MODIS visible images as well as elevated MODIS AOT and OMI SO<sub>2</sub>) are spatially and temporally co-located with the anomalous CO values and thus confirm their volcanic origin.

[21] We have ruled out a spurious origin for the anomalous CO values due to water vapor or aerosols in the volcanic plume. By modeling the effect of water vapor in MOPITT radiances we have shown that differences between water vapor observed in the plume versus the background could not account for the change in measured MOPITT radiance. Our analysis of IASI BT spectra indicates that the MOPITT TIR bandpass is not affected by aerosols in the volcanic plume. We have also shown that the depth of diagnostic CO lines increases as the distance to the volcanic plume decreases. Since the depth of these CO absorption lines should increase with increasing CO, we interpret this as definitive evidence that the high CO values retrieved in the plume are volcanic in origin.

[22] CO emissions result in an increase of CO<sub>2</sub>, CH<sub>4</sub>, and tropospheric O<sub>3</sub>, and thus have a positive indirect radiative forcing of approximately 0.2  $\text{W}/\text{m}^2$  (i.e., 12.5% of the total net anthropogenic forcing) [Forster *et al.*, 2007]. Global CO emissions measured in the 2002–2009 period range widely, between 1318 and 1504 Tg/year [Fortems-Cheiney *et al.*, 2011]; it is assumed that they are dominated by incomplete fossil fuel combustion and biomass burning. Fluctuations in gas emissions from natural sources (e.g., volcanic degassing) need to be quantified so as to separate them from those of anthropogenic origin.

[23] The magnitude of global volcanic CO emissions is unknown. Next, we estimate their approximate value to put them into perspective respect to other CO sources. Global volcanic CO<sub>2</sub> emissions have been estimated to be between 130 and 440 Tg/year (from data in Gerlach [2011]). Volcanic CO and CO<sub>2</sub> emissions depend on tectonic environment and may vary from eruption to eruption. However, and unlike CO and SO<sub>2</sub>, we find that CO and CO<sub>2</sub> are correlated to some degree, regardless of tectonic setting. Based on mean global volcanic CO<sub>2</sub> and linear regression parameters derived from volcanic CO and CO<sub>2</sub> measurements [Halmer *et al.*, 2002; Wardell *et al.*, 2004; Oppenheimer *et al.*, 2011] we estimate that global average volcanic CO emissions may be on the order of  $\sim 5.5$  Tg/year; errors from the linear regression are negligible compared to the CO<sub>2</sub> estimates. This is equivalent to the annual CO emissions produced by fuels (fossil and biofuel) in Australia [Kopacz *et al.*, 2010]. Though modest compared to anthropogenic sources, volcanic CO emissions are non-negligible.

[24] Satellite measurements of volcanic CO may lead to a better understanding of variations in the global CO budget, and thus improve climate models. They will also refine our understanding of volcanic processes by answering basic questions such as how much CO is emitted in an eruption

and what is the relative gas composition in different volcanic events.

[25] **Acknowledgments.** Thanks to David Edwards and Gene Francis for discussions on radiative effects and modeling help. Lieven Clarisse provided IASI BT spectra. Armin Rauthe-Schöch provided CARIBIC CO data. OMI retrievals were obtained with the Giovanni online data system, developed and maintained by the NASA GES DISC. IASI CO retrievals were performed at Université Libre de Bruxelles and can be downloaded from the French Ether atmospheric database (<http://ether.ipsl.jussieu.fr>). We appreciate the helpful comments from Anne Boynnard and Laura Pan. This paper benefited from reviews from Matthew Watson and Florian Schwandner. We acknowledge the mission scientists and Principal Investigators who provided the data used in this research effort. The NCAR MOPITT project is supported by NASA's Earth Observing System Program. NCAR is sponsored by the National Science Foundation.

[26] The editor thanks Iain Matthew Watson and Florian Schwandner for assistance evaluating this manuscript.

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