SO_2 atmospheric loading revealed through ground-based and satellite measurements.

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***Abstract*— Under the framework of the European Union Air Quality Monitoring and Forecasting in China[I], *Amfic*, project the sulfur dioxide, *SO_2*, columns from the Ozone Monitoring Instrument flying on board NASA's Aura satellite, *OMI/Aura*, and the *Sciamachy* instrument flying on board ESA's EnviSat satellite, have been compared to global ground-based measurements. The main objective of this work is to assess the ability of both satellite and ground-based measurements to reveal sources and transport mechanisms of anthropogenic SO_2 pollution on a global scale. The study commences with a detailed examination of each of the measurements separately, and then proceeds with a common revision. The strengths and shortcomings of each piece of information are identified, discussed and commended upon in turn in the following work. In the least, possible issues that might improve the measurements are to be identified and suggestions given.**

***Index Terms*— Atmospheric measurements, ultraviolet spectroscopy, air pollution.**

Manuscript received July, 10th, 2009. This work was supported by the European Union *Air Quality Monitoring and Forecasting in China* project.

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[I] http://www.amfic.eu/

I. Introduction

The main focus of this work is to assess the ability of satellite measurements to observe the sulfur dioxide, SO_2, content in the lower troposphere of anthropogenic origin. Sulfur dioxide anthropogenic sources are mainly coal/lignite/oil burning for electricity generation processes and contribute about a half of the sulfur loading in the atmosphere [1]. Monitoring of SO_2, pollution has been traditionally achieved through the air quality monitoring stations found mainly in urban and industrial areas, managed by the local authorities of each country or state, or via ground-based total ozone monitoring stations which also provide total SO_2 estimates [2], [3]. With the advancement of satellite instrumentation and technology it has become possible to monitor large events of SO_2 a loading in the atmosphere, such as volcanic outgassing and eruptions [4],[5] or large and localized sources of continuous SO_2 pollution [6]. In the present we wish to assess the ability of the synergistic use of ground-based and satellite measurements to depict and represent the background and daily anthropogenic SO_2 atmospheric loading. For this purpose, satellite measurements of the SO_2 atmospheric content are compared and discussed against two different satellite measurements. In the following, we first briefly present the datasets used and then discuss selected comparison results on the local and global scale.

II. Data sets

The ground-based observations used in this study are daily mean values of SO_2 total column in Dobson units, DU, extracted from Brewer spectrophotometers global measurements. The direct solar irradiance observations are used to derive the total ozone column using the Kerr algorithm [7] which is then applied again to the data in order to further extract the SO_2 total column. This procedure may lead to negative SO_2 total column values and even though these negative values have no physical meaning they are included in the data sets, since such values indicate very low total SO_2 levels. Daily data have been downloaded from the World Ozone and Ultraviolet Radiation Data Centre[II], *WOUDC*. Forty-three stations around the world provide daily SO_2 columns and associated error bars in the time frame of interest to this work.

The Scanning Imaging Absorption Spectrometer for Atmospheric CHartographY, SCIAMACHY, instrument has been flying on board ESA's EnviSat satellite since late 2003. The total SO_2 column is retrieved from the UV backscatter spectra by applying the Differential Optical Absorption Spectroscopy (DOAS) approach [8]. The interference between ozone and SO_2 is also present in this dataset and, as for the ground-based data discussed above, also leads of possible negative total SO_2 columns. The details of the DOAS analysis of the Sciamachy dataset can be found [9], [10] and [11]. For the purposes of this work, Sciamachy overpass files were created for the years 2005 to 2007 inclusive at a distance of 50km for the ground-based Brewer stations[III]. The satellite algorithm provides three total SO_2 columns depending on the postulated plume height, at 1km, 6km and 14km altitude. For the purposes of this air quality study, the low plume height measurements were taken into consideration. Only measurements flagged as completely successful retrievals, with a cloud fraction of less than 20% and a solar zenith angle, *SZA*, of less than 75° were used. The subset used here forms part of a more comprehensive dataset created for the Tropospheric Emission Monitoring Internet Service, TEMIS[IV] and the PROtocol MOniToring for the GMES Service Element, PROMOTE[V], projects.

The OMI SO_2 product is produced from global mode UV measurements of the Ozone Monitoring Instrument (OMI) on board the EOS Aura satellite [12], [13]. The data used in this study is distributed via the Aura Validation Data Centre as overpass files over selected regions around the world. For the purposes of this work, OMI overpass files were created from August of 2004 to 2008 inclusive, for a distance of 50km from the ground-based instruments[VI]. The total SO_2 column corresponding to the lowest assumed plume of 0.9 km was utilized, as for the Sciamachy data, and for a cloud fraction of less than 20%, SZA of less than 75° and OMI cross-track position between 10 and 50 to assure as near-nadir observations as possible.

III. Results

The comparisons between ground-based and satellite daily SO_2 columns were performed on different levels; firstly, each station was analyzed vis-à-vis the raw differences [in DU] between the two products, the monthly differences, the monthly time series and histogram representation in order to identify specific features of interest. Then, for the entire dataset, scatter plots for the summer and winter months were created alongside the seasonal variability, the dependence on the SZA and time series for the two hemispheres. A representative sample of this analysis is presented in the graphs below.

In Figure 1, upper graph, comparisons between ground-based and OMI total SO_2 columns are presented. In the upper graph, the time series of the monthly means for the station of Hohenpeissenberg is depicted in blue for the Brewer and in red for the OMI measurements. Hohenpeissenberg is a northern mid-latitude station at 1000m altitude, with clean air in its surroundings whose Brewer measurements are being regularly calibrated. The ground-based data show higher monthly mean values that the equivalent satellite estimates, as expected. For some seasons, as for the second part of year 2004, the seasonality is equally well depicted from either set of data, something that does not follow for the first half of year 2005. The interannual variability of the SO_2 loading even over a locally clean station such as this one is obvious from the ground-based data [in blue], and further testifies to the importance of transported pollution in the lower troposphere.

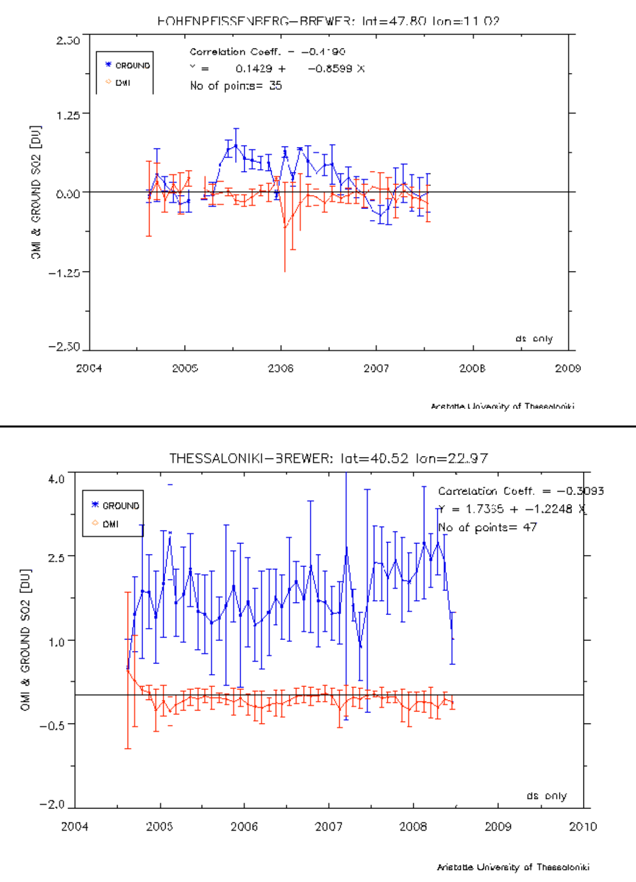

Figure 1. Upper. The monthly mean Brewer [in blue] and OMI [in red] atmospheric loading over Hohenpeissenberg. **Lower**. Same as above for the station of Thessaloniki.

In Figure 1, lower graph, the same is shown for the case of Thessaloniki. Thessaloniki is a sea-side metropolis in the North of Greece with both local and transported sources of

[II] http://www.woudc.org/
[III] http://www.oma.be/BIRA-IASB/Molecules/SO2archive/cases/china.php
[IV] http://www.temis.nl/
[V] http://www.gse-promote.org/
[VI] http://avdc.gsfc.nasa.gov/

SO_2 [14], [15]. The Brewer is being regularly calibrated and the ozone retrievals are in excellent agreement with the satellite data [16]. The high SO_2 values observed by the Brewer, attested by the mean difference of 1.80±1.40 D.U., compared to the OMI measurements, can be attributed to boundary layer pollution that the satellite is not able to capture [15].

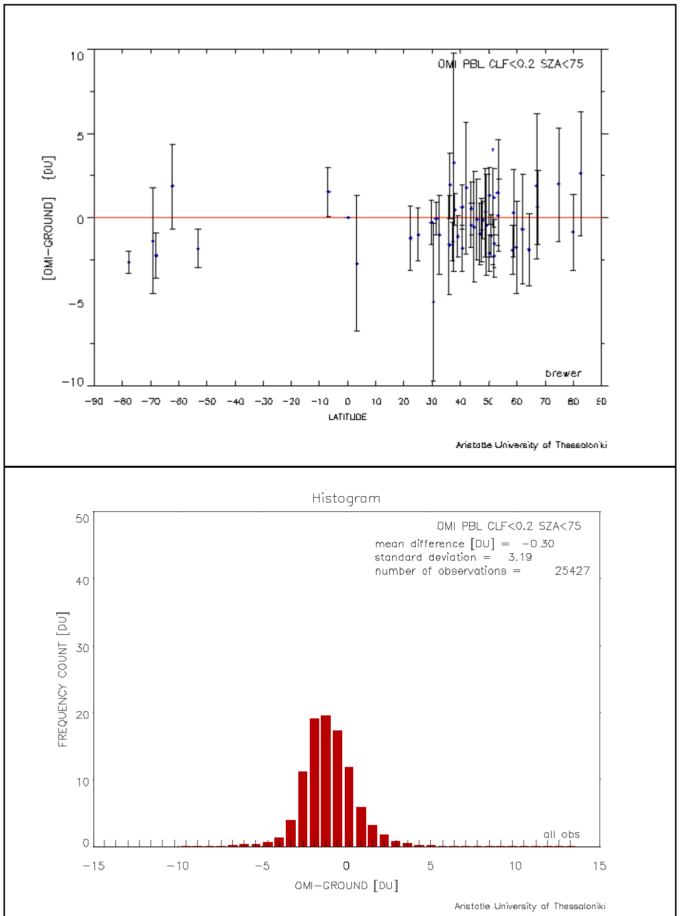

Figure 2. Upper. The global latitudinal dependence of the mean differences between Brewer and OMI. **Lower**. The histogram depiction of the daily absolute differences for all stations considered.

In Figure 2, comparisons between ground-based stations and OMI overpass assessments of the total SO_2 columns are presented for the totality of the stations we have analyzed. In the upper graph, the possible latitudinal dependence of the differences was assessed. Each of the stations is represented by a point in the graph. Apart from the obvious fact that the Northern mid-latitudes are well represented in this dataset compared to the Southern hemisphere in particular, no obvious dependency was found. In the lower graph, the histogram depiction of the absolute daily differences between ground and satellite are shown, with a mean satellite underestimation of 0.30 and quite large variance of 3.20 D.U.

In Figure 3, comparisons between ground-based and Sciamachy total SO_2 columns are presented. In the upper graph, the time series of the monthly means for the station of Hohenpeissenberg is depicted in blue for the Brewer and in red for the Sciamachy measurements. The monthly mean correlations are very good, especially from early 2006 onwards, where the seasonality of the SO_2 loading is observed similarly from ground and space. In the lower graph, the histogram depiction of the daily differences between Brewer and OMI are shown for the case of Thessaloniki. The double peak in the differences, which show a mean of 1.60±1.00 D.U., is attributed to the ground-based data. It should be noted here that, even though the Brewer is calibrated often, the extraction of the SO_2 signal depends heavily on the calibration parameters and small discontinuities in the database might introduce such artifacts in the comparisons.

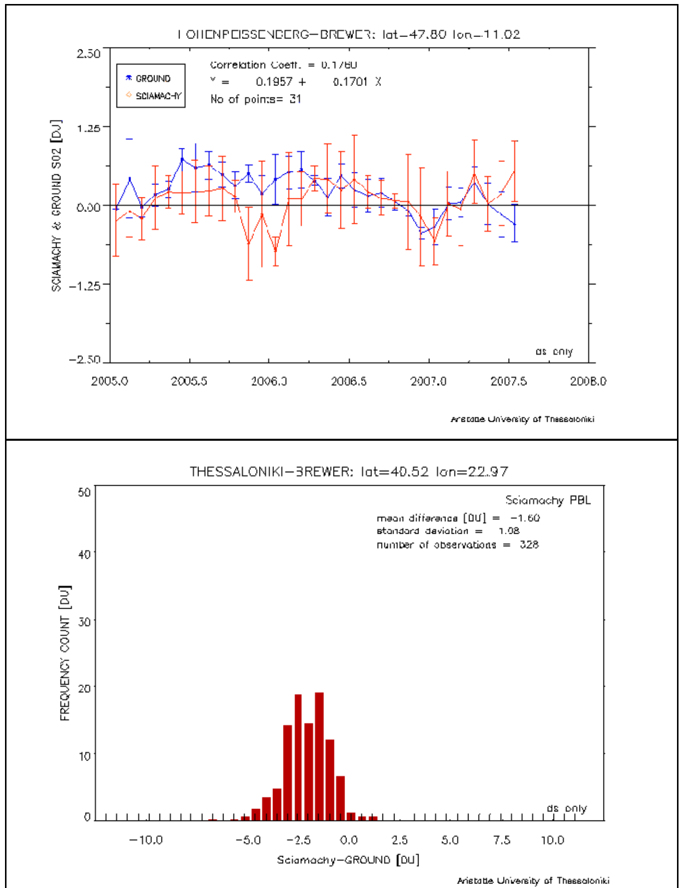

Figure 3. Upper. The mean Brewer [in blue] and Sciamachy [in red] atmospheric loading over Hohenpeissenberg. **Lower**. The histogram depiction of the absolute daily differences over Thessaloniki.

In Figure 4, comparisons between ground-based stations and Sciamachy overpass assessments of the total SO_2 columns are presented for the totality of the stations we have analyzed. As for the OMI comparisons, the northern mid-latitudes are favored in number of stations and show some structure, with some stations around 40°N overestimating the total SO_2 load. However, most stations underestimate the ground-based assessments. In the lower graph, the time series of the monthly mean observations for the Brewer [in blue] and Sciamachy [in red] measurements are shown for the entire dataset. The seasonality in the atmospheric SO_2 loading seems to be captured by both sets of instruments even though the amplitude differs, with the ground-based data showing higher values that the satellite counterparts.

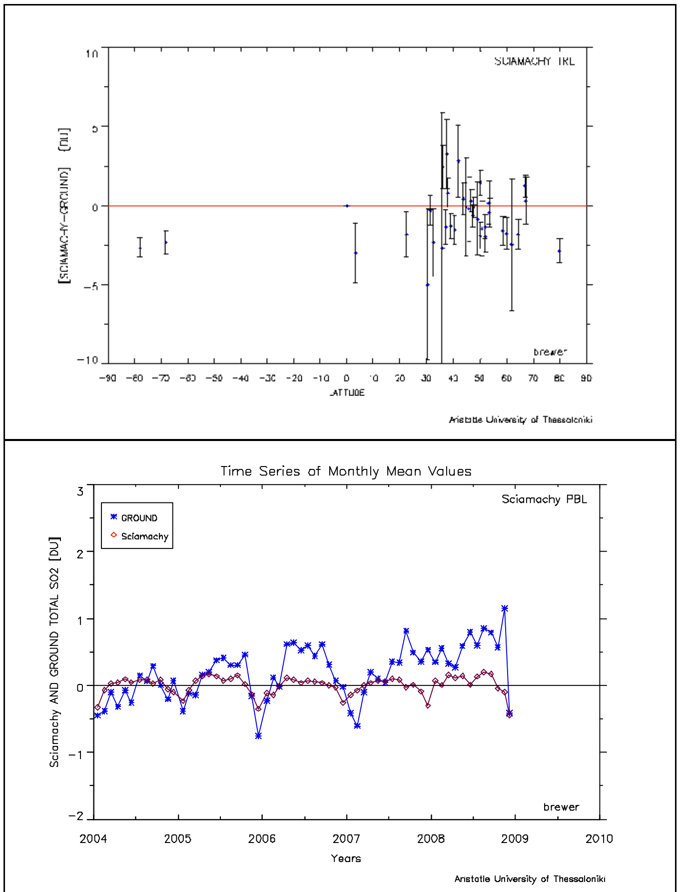

Figure 4. Upper. The global latitudinal dependence of the mean differences between Brewer and Sciamachy. **Lower**. The monthly mean time series of Sciamachy [in red] and Brewer [in blue] for all stations.

IV. Conclusions

Brewer ground-based measurements of the total SO_2 column at various sites around the world has been compared to both OMI/Aura and Sciamachy/EnviSat measurements of the planetary boundary layer SO_2 column, where most of the anthropogenic pollution resides. This study showed a high variability in the ground-based measurements, with values as high as 10 D.U. in various localities, whereas the satellite estimates range around the estimated noise level of 1 D.U. on average, with little apparent seasonality. The synergistic use of both sources of information reveals the shortcomings and strengths of each different type of measurement, and both used together.

Acknowledgements

We would like to acknowledge the World Ozone and Ultraviolet Radiation Data Centre, WOUDC, for the distribution of the Brewer spectrophotometer total SO_2 columns; NASA's Aura Validation Data Centre, for the distribution of the OMI total SO_2 overpass files; the Tropospheric Emission Monitoring Internet Service, TEMIS and the PROtocol MOniToring for the GMES Service Element, PROMOTE, projects for the Sciamachy SO_2 overpass files. This work was performed under the auspices of the European Union Air Quality Monitoring and Forecasting in China, Amfic, project.

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