MarcoPolo

Monitoring and Assessment of Regional air quality in China using space Observations.

Project Of Long-term sino-european co-Operation

Type of funding scheme: Collaborative Project - Small or medium-scale focused research project
Work programme topics addressed: SPA.2013.3.2-01: Cooperation with third countries

**Deliverable D3.4 (PU)**

< SO$_2$ Emissions Estimates over China>

Version: 1.0

**Due date of final deliverable:** project month 24  
**Actual submission date:** project month 34

**Organisation name of lead contractor for this deliverable:** Aristotle University of Thessaloniki
## Document Status sheet

<table>
<thead>
<tr>
<th><strong>Lead Authors</strong></th>
<th>MariLiza Koukouli</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Contributing Authors</strong></td>
<td>Dimitris Balis</td>
</tr>
<tr>
<td><strong>Reviewed by</strong></td>
<td>Ronald van der A</td>
</tr>
<tr>
<td><strong>Distribution list</strong></td>
<td>Project partners and EC</td>
</tr>
</tbody>
</table>

## Document Change Record

<table>
<thead>
<tr>
<th>Date</th>
<th>Issue</th>
<th>Pages affected</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Friday 07 October 2016</td>
<td>1.0</td>
<td>all</td>
<td>&lt;First version of the report initialised&gt;</td>
</tr>
</tbody>
</table>
Table of Contents

1 Introduction ........................................................................................................................................... 4
2 Data and Methodology .......................................................................................................................... 4
  2.1 The MEIC emission inventory ........................................................................................................ 4
  2.2 The OMI/Aura SO₂ observations ................................................................................................... 5
  2.3 The CHIMERE model SO₂ output ................................................................................................ 6
3 Results .................................................................................................................................................. 8
  3.1 Calculating the top-down emission inventory................................................................................... 8
  3.2 Calculating the aposteriori emission inventory............................................................................... 9
4 Conclusions ......................................................................................................................................... 10
Description of emission inventory provided to MarcoPolo ....................................................................... 11
Appendices ........................................................................................................................................... 12
References ............................................................................................................................................. 18
Acknowledgments ................................................................................................................................. 19
1 Introduction

With the rapid development of the Chinese economy since 2000, sulfur dioxide, SO$_2$, emissions from China have been of increasing global concern. There have been indications that the emission growth rate slowed around 2005 and that emissions began to decrease after 2006, mainly due to the wide application of flue-gas desulfurization devices in power plants. However due to the differences in growth rate among the many Chinese Provinces, it remains to be seen whether this decreasing trend can be verified for the entire domain. In this study we use space-based observations of SO$_2$ columns from the Ozone Monitoring Instrument on board the Aura satellite, OMI/Aura, to derive monthly top-down SO$_2$ emissions over China via inverse modeling with the CHIMERE chemical transport model. The driving SO$_2$ emissions have been provided by the Multi-resolution Emission Inventory for China (MEIC) model. In MEIC, the SO$_2$ sources are further partitioned among industry, power and residential provenances. This study aims at providing updated SO$_2$ emissions, based on satellite observations, which may later be used to update existing chemical transport model results.

During the past decade or so, with the rapid improvement of satellite instrumentation as well as novel algorithmic development, the monitoring of atmospheric pollutant emissions on a global scale has shifted towards the satellite community. Especially China, with its rapid economic growth, became a focal point of many studies, especially for the NOx family of gases [e.g. Zhang et al., 2007; Gu et al., 2014]. In this framework the estimation of sulphur dioxide, SO$_2$, emissions over China, as provided by satellite observations is attempted. The methodology followed, discussed in Section 3, requires an existing, apriori, emission inventory [presented in Section 2.1], chemistry-transport modelling of said emissions in order to extract an SO$_2$ modelled field [Section 2.2] and the satellite SO$_2$ observations [Section 2.3.] The calculated top-down emission field is presented and discussed in Section 3.1 whereas the final, aposteriori, updated emission inventory is shown in Section 3.2.

2 Data and Methodology

2.1 The MEIC emission inventory

The Multi-resolution Emission Inventory for China, MEIC, compiled by Tsinghua University, Beijing, China, is a production-based inventory, updated from the widely used INTEX-B data set (Zhang et al., 2009). The inventory covers 31 provinces or autonomous regions, 10 pollutants, such as SO$_2$, NOx, CO, PM 2.5, PM10, ammonia, organic
carbon, etc., and 700 emission source categories. In this study, the 2010 SO$_2$ emission inventory has been extracted from the http://www.meicmodel.org repository on a monthly basis and a 0.25°x0.25° grid. The SO$_2$ emissions, in Mg/month, are provided for four sectors: power, industry, residential, and transportation. The domain studied here spans from 102°E to 132°E and from 15°N to 55°N. In Fig. 1, the emission separated in the four sectors are given for the month of August 2013, with industry and power [first row] producing the highest yields, followed by residential consumption [bottom left] and transportation [bottom right.]

![MEIC Emissions | SO$_2$ Mg/Month](image)

Fig. 1. The MEIC emission inventory for August 2010 in Mg per month for four sectors: upper left, industry; upper right, power; lower left, residential and lower right, transportation. Please note the different colour scales.

### 2.2 The OMI/Aura SO$_2$ observations

Daily OMI/Aura planetary boundary layer SO$_2$ observations for year 2013 and the same domain discussed above have been provided by the Belgian Institute of Aeronomie, BIRA, algorithm [Theys et al., 2015]. OMI is a nadir, UV-Vis-viewing instrument, on board the Aura satellite with a local solar overpass time of 13:30L.T. and a 13 km x 25
km pixel size [Levlt, et al., 2006]. The retrieval of SO$_2$ vertical column densities, VCDs, using the BIRA algorithm is achieved by applying the Differential Optical Absorption Spectroscopy technique to the measured spectra in the 312–326 nm wavelength range.

The observations were filtered for high solar zenith angles over 70°, high cloud fractions over 20% and, using robust algorithm flagging, were averaged onto a 0.25°x0.25° monthly grid using a 0.75° smoothing average box. In Fig. 2, left, an example of the month of August 2013 is shown where the SO$_2$ load in Dobson Units, D.U., is given.

![Image of SO$_2$ load in Dobson Units for August 2013](image.png)

**Fig. 2.** Left. The OMI/Aura area weighted gridded mean SO$_2$ observations for August 2013 in D.U. Right: The CHIMERE model SO$_2$ output for August 2013 in D.U. based on the MEIC emissions shown in Fig. 1.

### 2.3 The CHIMERE model SO$_2$ output

CHIMERE is an Eulerian off-line chemistry-transport model, CTM (Menut et al., 2013). External forcings are required to run a simulation: meteorological fields, primary pollutant emissions and chemical boundary conditions. Using these input data, CHIMERE calculates and provides the atmospheric concentrations of tens of gas-phase and aerosol species over local to continental domains (from 1 km to 1 degree resolution). In this work, the CHIMERE model version v2013b was applied to the domain 102°E - 132°E and 15°N - 55°N for year 2013 for a 0.25°x0.25°
resolution. The anthropogenic emission inventory used in this CHIMERE run is the MEIC inventory discussed in Section 2.1, whereas the biogenic emission results from the Model of Emissions of Gases and Aerosols from Nature, MEGANv2.1, dataset (Guenther et al., 2012.) The meteorological input has been extracted from the European Centre for Medium-Range Weather Forecasts, ECMWF, operational data. The output SO$_2$ profile is given in 8 vertical levels, from surface up to 500hPa, which were integrated using the standard atmospheric equations to produce SO$_2$ columns in D.U. The month of August 2013 is shown as example in Fig. 2, right.

Fig. 3. The top-down emission method applied to the month of August 2013. Upper left, the OMI SO$_2$ load in D.U.; upper right, the CHIMERE SO$_2$ model output in D.U.; lower left, the MEIC apriori SO$_2$ emissions and lower right in Mg/Month, the top-down SO$_2$ emission inventory in Mg/Month.
3 Results

3.1 Calculating the top-down emission inventory

\[ E_t = E_a \frac{\Omega_t}{\Omega_a} \]  \hspace{1cm} \text{Equation 1}

Following the mass balance method (Leue et al., 2001) as described by Martin et al. (2003 and 2006), the top-down SO\(_2\) emission inventory, \( E_t \), may be calculated as a function of the apriori emission inventory, \( E_a \), a CTM SO\(_2\) field, \( \Omega_a \), based on the same apriori emissions and the satellite SO\(_2\) observations, \( \Omega_t \), as shown in Eq. 1. In our case, \( E_a \) is provided by the MEIC emission inventory [Section 2.1], \( \Omega_a \) by the CHIMERE model output [Section 2.3] and \( \Omega_t \) by the OMI/Aura BIRA data [Section 2.2]. The associated top-down SO\(_2\) emission inventory error, \( \varepsilon_t \), is calculated according to Eq. 2, where, \( \varepsilon_a \) denotes the error in the apriori emission field, \( E_a \), assumed here to be 50% of the original emissions and \( \varepsilon_{\Omega t} \) denotes the error in the OMI SO\(_2\) observations calculated as discussed in Theys et al., 2015.

\[ \varepsilon_t^2 = \left( \frac{\Omega_t}{\Omega_a} * \varepsilon_a \right)^2 + \left( \frac{E_a}{\Omega_a} * \varepsilon_{\Omega t} \right)^2 \]  \hspace{1cm} \text{Equation 2}

The top-down emission inventory is given in the bottom left panel of Fig. 3 in Mg/month [bottom right] for the month of August 2010. The OMI SO\(_2\) observations are shown in the upper left, where any white areas within the domain studied denote areas of no satellite observations for that month. In the upper right, the CHIMERE SO\(_2\) model output is presented, where areas in purple denote near-zero SO\(_2\) concentration. In the bottom left, the MEIC apriori SO\(_2\) emissions are shown for all four sectors totaled. As expected, the top-down emissions follow closely the patterns of the apriori emissions albeit with higher absolute levels and also high emissions in regions where the satellite observations note a strong SO\(_2\) source and the CHIMERE model was not able to capture that load.
3.2 Calculating the aposteriori emission inventory

The calculated \textit{top-down} emission inventory, $E_t$, may be combined with the \textit{apriori} emission inventory, $E_a$, to provide an \textit{aposteriori} emission inventory, $E_p$, following the maximum likelihood theory and a log-normal distribution of errors. In Equation 3 the calculation of the \textit{aposteriori} emission inventory is given, and its associated errors in. Equation 4. Hence, in this methodology, the original bottom-up emission inventory is integrated with the top-down satellite observations, weighted by their respective errors, and using modeling outputs as background field, in order to constraint, update and provide new emissions estimates. It also follows that since the \textit{apriori} emission field is weighted by the top-down emission field error, and vice versa, the \textit{aposteriori} will depend mostly on the \textit{apriori} should the errors of the top-down be too large, and vice versa. In that way, it is assured that at locations where the satellite observations are too sparse or the information content in the SO$_2$ load too low, the \textit{aposteriori} emission field will revert back to the \textit{apriori}.

\[
\ln E_p = \frac{\ln E_a (\ln \varepsilon_t)^2 + \ln E_t (\ln \varepsilon_a)^2}{(\ln \varepsilon_t)^2 + (\ln \varepsilon_a)^2}
\]

\textit{Equation 3}

\[
(\ln \varepsilon_p)^{-2} = (\ln \varepsilon_t)^{-2} + (\ln \varepsilon_a)^{-2}
\]

\textit{Equation 4}

An example of the \textit{aposteriori emissions} calculated with the methodology and input parameters discussed above is shown in Fig. 4. For the month of August 2010, the original, bottom-up, MEIC emission field is shown in the top left, whereas the \textit{aposteriori} emissions in the top right. The percentage differences and their absolute differences are shown in the bottom row, left and right, respectively.
Fig. 4. The original, bottom-up, MEIC emissions [upper left] compared to the aposteriori emissions calculated in this work [upper right] are shown for August 2010 in Mg/month. The percentage differences and the absolute differences are shown in the bottom row, left and right, respectively.

4 Conclusions

In this work, the first step towards creating a satellite-based emission inventory for the SO$_2$ load over China has been presented using the month of August 2010 as example and input from the CHIMERE CTM, the MEIC emission inventory and the OMI/Aura observations. The technique applied has already been verified in previous works,
mainly for NOx, and is furthered into calculating an *aposteriori* emission inventory which may then be used as input to a CTM run in lieu of the *apriori* emission field.

**Description of emission inventory provided to MarcoPolo**

The *aposteriori* SO\(_2\) emissions for year 2014, in the domain from 102°E to 132°E and from 15°N to 55°N, in a 0.25°x0.25° spatial resolution and monthly temporal resolution, have been provided to the MarcoPolo project and can be found at [http://www.marcopolopanda.eu/wp/data/inventory/MarcoPoloInventory_v0.1.zip](http://www.marcopolopanda.eu/wp/data/inventory/MarcoPoloInventory_v0.1.zip).

The netcdf data files contain the following structure:

- **Dimensions**
  - lat = 129
  - lon = 121
- **Attributes**
  - author = "MariLiza Koukouli"
  - contact information = "mariliza@auth.gr"
  - institution = "Laboratory of Atmospheric Physics, Aristotle University of Thessaloniki"
  - time frame = "2014"
  - sector classification = "total emissions"
  - emis_cat_name = "sulphur dioxide emissions"
  - source_type_name = "sulphur dioxide emissions"
  - pollutant_description = "updated sulphur dioxide emissions based on the CHIMERE model running the MEIC emissions and the OMI/Aura observations"
  - unit_emissions = "Mg/month"
  - nodata_value = "-9999.0"
- **Variables**
  - float emissions(lon, lat)

In Appendix I and Appendix II the emission inventory is presented for the full twelve months of 2014.
Appendices.

Appendix I. The monthly emission fields as calculated in this work over China for year 2014. From left to right and top to bottom, the twelve months of the year are shown. In each panel: top left, the OMI/Aura SO\(_2\) load in D.U.; top right, the CHIMERE SO\(_2\) load in D.U.; bottom left, the aposteriori emissions in Mg/month and in the bottom right, the difference to the apriori MEIC emissions in %.
Appendix II. The monthly emission fields as calculated in this work over China for year 2014. From left to right and top to bottom, the twelve months of the year are shown. In each panel: top left, the MEIC apriori emissions in Mg/month; top right, the aposteriori emissions in Mg/month; bottom left, the difference between emission inventories in Mg/month and in the bottom right, the difference in %.
September 2014
MEIC Apriori Emissions [Mg/month]

October 2014
MEIC Apriori Emissions [Mg/month]

November 2014
MEIC Apriori Emissions [Mg/month]

December 2014
MEIC Apriori Emissions [Mg/month]
References


Acknowledgments

Results presented in this work have been produced using the European Grid Infrastructure (EGI) through the National Grid Infrastructures NGI_GRNET (HellasGrid) as part of the SEE Virtual Organization. The authors would like to acknowledge the support provided by the Scientific Computing Office, IT A.U.Th. throughout the progress of this research work.