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Short Communication

A new method for deriving trace gas emission inventories from satellite observations: The case of SO_2 over China

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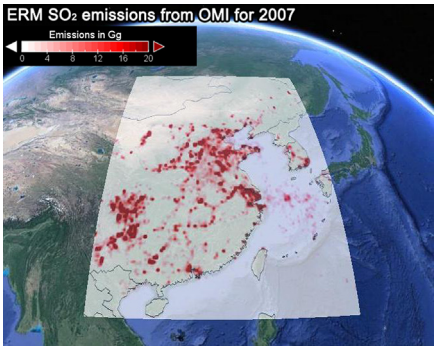
HIGHLIGHTS

- A new method (ERM) for deriving anthropogenic emissions from satellite data
- ERM is applied for SO_2 over China (2007–2011).
- The ERM-derived emissions agree well with bottom-up inventories.
- The lowest emissions are calculated for 2009 (21.7 Tg/yr).
- The highest emissions are calculated for 2007 (30.9 Tg/yr).

GRAPHICAL ABSTRACT



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ABSTRACT

A method is developed that allows the construction of spatial emission inventories. The method is applied for anthropogenic SO_2 over China (0.25° × 0.25°). The Enhancement Ratio Method (ERM) allows for the calculation of SO_2 emissions using relationships between gridded satellite measurements of SO_2 and NO_2 at low wind speeds, and satellite-based NOx emission estimates. Here, we derive SO_2 emissions for five years (2007–2011). A large decrease of emissions during 2007–2009 and a modest increase between 2010 and 2011 is observed. The evolution of emissions over time calculated here is in general agreement with bottom-up inventories, although differences exist, not only between the current inventory and other inventories but also among the bottom up inventories themselves. The ERM-derived emissions are consistent, spatially and temporally, with existing inventories.

1. Introduction

Air quality management relies on Chemistry-Transport-Models (CTMs) whose accuracy is to a large part dependant on the accuracy of emission inventories of primary pollutants (Zhao et al., 2011 and references therein). The more common way to construct an inventory is

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the so-called bottom-up compilation. Briefly, this involves statistics of emitting activities of various activity sectors (traffic, industry, residential, power generation) and associated typical activity unit emission factors. Most steps in the compilation process are characterized by large errors and uncertainties (Zhao et al., 2011; Hong et al., 2017). Additionally, the process requires a level of effort that makes frequent revisions unpractical (Mijling and van der A, 2012).

Emission inventories calculated from satellite data have advantages as compared to conventional bottom-up approaches (e.g. Streets et al., 2013). First of all, they are spatially consistent as the total emissions (satellite-based methods return total emissions only in contrast to bottom-up approaches) are calculated over all grid cells with nearly the same accuracy. Bottom-up inventories on the other hand may have varying accuracies over different grid cells depending on the sector apportionment over each cell and differences in the accuracy in determining the emission factors per sector. Additionally, they can be updated more frequently, more easily and using fewer resources than bottom-up inventories (e.g. Mijling and van der A, 2012). For southeast Asia, where current rapid economic evolution results in changes in emissions far more pronounced than in the rest of the world, these advantages are even more important (e.g. van der A et al., 2008; Koukouli et al., 2016). Given the time and effort required to compile bottom-up inventories, satellite observations can provide an important alternative approach (see Li et al., 2010; Lin et al., 2010; Liu et al., 2015, 2017). Because satellites measure atmospheric concentrations and not emissions, inverse modelling techniques need to be used alongside. Briefly, they involve the use of CTMs to calculate a concentration field from a certain emission inventory. The modelled concentrations are then compared to the satellite concentration field and this information is used for adjusting the a-priori emissions (see, e.g., Sandu and Chai, 2011; Streets et al., 2013 for more info).

During the last few years, a variety of published studies estimated NOx emissions from satellite data. These studies applied various techniques, depending on the required resolution in time and space (e.g. Mijling and van der A, 2012 and references therein). A few studies have used satellite data to calculate NOx emissions from megacities (Beirle et al., 2011). Also, recently, SO_2 emissions from point sources have been successfully calculated using satellite-based observations (Fioletov et al., 2011, 2013, 2015, 2016).

Few spatial inventories have been calculated from satellite data for atmospheric constituents other than NOx such as CH_4 and non-methane volatile organic compounds (e.g. Frankenberg et al., 2005; Stavrakou et al., 2009). Apart from this fact, spatial emission inventories compiled from satellite observations require the use of a CTM. In this work, a method that can calculate spatial inventories of trace gases from satellite data and requires the a-priori involvement of a NOx emission inventory only is presented for the first time. This method is applied for the derivation of an SO_2 emission inventory on an annual basis for China. It has to be highlighted here that this short communication paper is dedicated to the presentation and evaluation of the method and not to an in-depth analysis of SO_2 emission variabilities over China.

2. Data and method

Here, daily SO_2 and NO_2 data at a spatial resolution of 0.25° × 0.25° from the OMI instrument on the EOS Aura satellite are utilized. The data span from 2007 to 2011. Specifically, tropospheric NO_2 concentration data in 10^{15} molecules/cm^2 (OMNO2d v2.1) (Bucsela et al., 2013; Krotkov, 2013) and planetary boundary layer (PBL) SO_2 column data in Dobson Units (OMSO2e v1.1.7) (Krotkov et al., 2012; Li et al., 2013) are analyzed. The OMNO2d gridded data include clear sky retrievals (cloud fraction < 30%) while the OMSO2e gridded data are produced from best level-2 pixel data, screened for OMI/Aura row anomalies and other data quality flags. The SO_2 data are brought to the same concentration units like NO_2 (10^{15} molecules/cm^2) prior to their use. Additionally, daily gridded 0.25° × 0.25° 10 m wind speed (ws) data from the ERA-Interim Reanalysis (Dee et al., 2011) are used. The data are produced by ECMWF (European Centre for Medium-Range Weather Forecasts) Integrated Forecast System (IFS) at a native horizontal resolution of ~79 km assimilating satellite and in-situ observations and can be acquired at various resolutions (here 0.25° × 0.25°) from ECMWF's website. The NOx emission data are from KNMI's DECSO algorithm (Daily Emission estimates Constrained by Satellite Observations) (Mijling and van der A, 2012; Mijling et al., 2013). The algorithm uses daily columnar concentrations from satellites to estimate emissions of short-lived atmospheric constituents on a mesoscopic scale. It involves the use of a forward model run from the CHIMERE CTM, trajectory analysis to take into account the transport away from the source and the use of a Kalman filter for the inversion (see Mijling and van der A, 2012 for more details). Here, DECSO v1 and v3a monthly data are utilized, which are constructed using OMI/AURA measurements from the KNMI product v2 (Boersma et al., 2011). DECSO v3a is a more recent development of v1, with some improvements in the concentration simulation, the source-receptor calculation and the inversion algorithm, the latter featuring full inversion in the Kalman gain matrix calculation. It also features emission injection height according to sector (see Ding et al., 2015 for more details on v3a).

The slope of the linear regression curve between measured SO_2 and measured NOx equals to the enhancement ratio (ER) (Kourtidis et al., 1999).

$$\mathrm{ER} = \frac{\Delta[\mathrm{SO_2}]}{\Delta[\mathrm{NOx}]} \tag{1}$$

In the ideal case of a box with no transport in or out and no chemical or physical processes of losses/additions, it is obvious that the ratio of the concentrations of two substances will be equal to their emission ratio. In reality, while for many species it is true that no additions due to chemical or physical processes occur within the box, it is also true that within each grid box, emissions of species occur simultaneously with their transport out of the box and transport into the box from neighbour cells. Transport out of the area will affect both substances in the same manner. This is because:

$$\mathrm{T(out)_{species}} = \mathrm{function(ws \cdot [species])} \tag{2}$$

where $\mathrm{T(out)_{species}}$ is the amount of the species transported out of the area, ws is the near surface wind speed and [species] is the concentration of the species. Thus, for two species 1 and 2, their resulting concentration ratio will be:

$$\begin{aligned}\frac{[\mathrm{species1_{final}}]}{[\mathrm{species2_{final}}]} &= \frac{[\mathrm{species1_{initial}}]-\mathrm{T(out)_{species1}}}{[\mathrm{species2_{initial}}]-\mathrm{T(out)_{species2}}}\\ &= \frac{[\mathrm{species1_{initial}}]-\mathrm{function(ws \cdot [\,species1_{initial}])}}{[\mathrm{species2_{initial}}]-\mathrm{function(ws \cdot [species2_{initial}])}}\\ &= \frac{c \cdot [\mathrm{species1_{initial}}]}{c \cdot [\mathrm{species2_{initial}}]} = \frac{[\mathrm{species1_{initial}}]}{[\mathrm{species2_{initial}}]}\end{aligned} \tag{3}$$

where c is the remaining fraction. Hence, it will still correspond to their emission ratio. Transport in the area will affect this relationship only when the concentrations ratio outside the area is very different from the one inside. In this case, the axis intercepts might change whereas the slope of the relationship will be only slightly affected. Chemical and physical losses also occur within the grid box, but if the source strength of the compounds involved in the calculations is high enough, the magnitude of these processes will be small compared to the emission rate of the compounds. These conditions can be largely met if only measurements are considered that were done under conditions that allow for little horizontal exchange in the grid box, i.e. under low