GlobEmission
ESA Contract No 4000104001/11/I-NB

Algorithm Theoretical Baseline Document
Version 3

Date of issue: 17 March 2015
<table>
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<td>Public</td>
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### Document Change Record

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<th>Date</th>
<th>Issue</th>
<th>Pages affected</th>
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<td>9 January 2012</td>
<td>draft</td>
<td>All</td>
<td>Template</td>
</tr>
<tr>
<td>4 April 2012</td>
<td>1.0</td>
<td>All</td>
<td>First issue</td>
</tr>
<tr>
<td>30 October 2012</td>
<td>2.0</td>
<td>52</td>
<td>Small updates</td>
</tr>
<tr>
<td>20 December 2013</td>
<td>2.2</td>
<td>52-54</td>
<td>Description new algorithm for spatial disaggregation</td>
</tr>
<tr>
<td>16 September 2014</td>
<td>3.0</td>
<td>All</td>
<td>New version for CCN1</td>
</tr>
<tr>
<td>10 December 2014</td>
<td>3.1</td>
<td>All</td>
<td>Updates</td>
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<td>17 March 2015</td>
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1 Introduction

1.1 Purpose and scope

The algorithms used in the services of GlobEmission to derive emission estimates from satellite data are described in detail in this document. Each of the 5 algorithms described has been developed for a specific type of emission and spatial scale. The performance of the algorithms in combination with the available satellite data has been tested. This document contains for each algorithm a description of the input data, detailed algorithm description and an error analysis. The error analysis is based on theoretical calculations but tested with real satellite data.

2 Documents

2.1 Reference documents

Chapter 4.1


**Chapter 4.2**


Chapter 4.3


Chapter 4.4


[RD-3] ESA Aerosol CCI - European Space Agency Aerosol Climate Change Initiative (http://www.esa-aerosol-cci.org/)


[RD-16] Shi Y., J. Zhang, J. S. Reid, B. Holben, E. J. Hyer, and C. Curtis, An analysis of the collection 5 MODIS over-ocean aerosol optical depth product for its implication in aerosol assimilation,

2.2 Informative documents

[ID-1] User Requirements Document (GE_URD_02_00)
[ID-2] Product Specification Document (GE_PSD_01_02)
3 Overview of algorithms

Satellite instruments measure high resolution radiance spectra at wavelengths in the ultraviolet, visible or infrared, which provide valuable information on the chemical composition of the atmosphere for various trace gases and aerosols. From their sun-synchronous orbit at a height of ~800 km they have a global coverage within 1-5 days at a 10-50 km resolution. The homogeneity and global coverage of the satellite retrievals are ideal to estimate top-down emissions. To achieve this, the retrieved tropospheric column concentrations of a trace gas are compared with the simulated concentrations from a chemistry transport model, based on a bottom-up emission inventory. The difference between observed and modelled concentrations contains information on how to adjust the underlying trace gas emissions. This is an inverse problem which is computationally challenging because the non-local relation (sensitivity) between emission and concentration has to be found. Due to transport away from the source, life-time information of the pollutant is crucial.

Several approaches have been developed, which are applied to different time scales and emission inventory resolutions. Shorter assimilation intervals ask for fast data assimilation algorithms, while transport issues become important for high spatial resolution. The methods differ in their emission domain (global or regional), the used satellite data, and the used chemical transport model, and sensitivity derivation.

Various techniques are used: e.g. adjoint modelling, Kalman filters, ensemble Kalman filter and 4D-VAR data assimilation. Also the chemical-transport models vary: IMAGES, CHIMERE, LOTOS-EUROS, SILAM. The details of the algorithms used in the GlobEmission project are discussed in detail in section 4.
4 Algorithm descriptions

4.1 Global Emission Estimates

4.1.1 Introduction

The adjoint model technique coupled with the IMAGESv2 global CTM is used for the derivation of satellite-based global emission estimates. The algorithm has been fully described in the first version of the ATBD document of GlobEmission (ATBD_02_01) (Figure 1). Below we give an overview of the input data used in the source inversion exercises (Section 1.1.2), and discuss model features regarding the HCHO and SO\textsubscript{2} model simulation (Section 1.1.3). Sensitivity inversions allowing to characterize uncertainties in the emission estimates are also presented (Section 1.1.3).

![Figure 1: A schematic view of the source inversion algorithm as implemented in IMAGESv2.](image)

4.1.2 Input data

**Satellite data:** HCHO vertical column abundances from the OMI and GOME-2 sensors. This dataset is obtained from the UV-Vis retrieval team of BIRA-IASB and is available at http://h2co.aeronomie.be. A new dataset of SO\textsubscript{2} column densities from the OMI instrument has been developed at BIRA-IASB and will be used to constrain the SO\textsubscript{2} source.
A priori emissions: Anthropogenic emissions are obtained from the RETRO 2000 database (http://retro.enes.org, Schultz et al., 2008), except over Asia where REASv2 database for year 2008 is used (Kurokawa et al., 2013). Open fires are provided by the Global Fire Emission Database (GFEDv3, van der Werf et al., 2010) and the emission factors are from the 2011 update of the database of Andreae and Merlet(2001). Vertical smoke profiles from open fires are obtained from Sofiev et al.(2013) and are based on plume top heights and MODIS fire radiative power. On average and on the global scale, half of emitted flux is injected in the boundary layer. The injection profile maps are obtained from the GlobEmission web portal (http://www.globemission.eu/data.php) and implemented in IMAGESv2. The diurnal cycle fire profile was derived based on Roberts et al.(2009). The global GFEDv3 NMVOC emission estimate is estimated at 105.4 TgVOC in 2010. Biogenic emissions of isoprene are obtained from the MEGAN-MOHVCAN inventory (http://tropo.aeronomie.be/models/isoprene.htm, Stavrakou et al., 2014). The global annual estimate of isoprene emissions is 363 Tg for 2010. The biogenic source of methanol is in Stavrakou et al. Stavrakou et al. (2011).

Anthropogenic SO₂ emissions are obtained from the REASv2 inventory (Kurokawa et al.2013) over Asia, from EMEP over Europe (http://www.ceip.at) and from the EDGAR3.2 FT2000 inventory for 2000 over the rest of the world. The global emissions of SO₂ from anthropogenic sources are estimated at 53.8 TgS in 2010. Vegetation fires are obtained from the GFEDv3 database and account for 1.3 TgS in 2010. Emissions of SO₂ and other sulfur compounds from continuously degassing volcanoes are constant and amount to 7.2 TgS annually (Andres and Kasgnoc, 1998). Besides direct emissions, SO₂ is formed through oxidation of sulfur-containing biospheric compounds like dimethyl sulfide (DMS), carbonyl sulfide (OCS), carbon disulfide (CS₂), and hydrogen sulfide (H₂S). The global annual photochemical source is calculated at 18.1 TgS. Dry and wet deposition account for approx. 60% of the global SO₂ sink, followed by oxidation by OH (20%), and heterogeneous in-cloud reactions of SO₂ with H₂O₂ (18%) and with O₃. The aerosol simulation in IMAGESv2 is described in Stavrakou et al.(2013).

4.1.3 HCHO simulation using the IMAGESv2 global CTM

IMAGESv2 global model provides the global distribution of about 130 chemical constituents between the Earth’s surface and 22.5 km, at a resolution of 2 degrees in latitude, 2.5 degrees in longitude and 40 vertically discretized levels. Advection is driven by monthly mean operational ECMWF ERA-Interim fields, and daily fields are used for temperature, water vapour, boundary layer mixing, and cloud optical depths. As the model time step is one day, diurnal variations in the photorates and in the concentrations are accounted for through correction factors computed via a diurnal cycle simulation with a 20-minute time step. The diurnal profiles are used to estimate the formaldehyde and SO₂ model columns at the overpass times of the two satellites (9:30 LT for GOME-2, and 13:30 LT for OMI) from the daily averaged values calculated with a time step of one day. The modelled columns are confronted with HCHO and SO₂ column data (binned onto the model horizontal grid and monthly averaged), following a 4-month spin-up time. The simulated monthly averaged
columns account for the vertical sensitivity of the measurements through the averaging kernels, and for the sampling times of observations at each location.

The inorganic chemistry reactions, as well as the methane degradation mechanism, are as in Müller and Stavrakou (2005), with updated kinetic rates from Sander et al. (2011). The degradation mechanism for the majority of the NMVOCs is largely based on the Master Chemical Mechanism (MCM) (Saunders et al., 2003). The isoprene oxidation chemistry follows the LIM0 mechanism (Peeters et al., 2014), which recycles OH more efficiently than generally assumed in models under low NOx conditions, (Peeters and Müller, 2010).

4.1.3.1 HCHO yields from the oxidation of pyrogenic NMVOCs

The oxidation chemistry of pyrogenic NMVOCs is presented in Stavrakou et al. (2009). According to this study, based on the quasi-explicit Master Chemical Mechanism v3.1 (Saunders et al., 2003), we performed time-dependent simulations under high NOx conditions using the chemical solver of the KPP package (Damian et al., 2002; Sandu and Sander, 2006). Simulations start at 06:00 for a temperature of 298K at a latitude of 30 degrees in February. The model is initialized with 0.1 ppb of the considered NMVOC, 35 ppb O3, and 100 ppb CO. The NO2 concentration is kept constant and is taken equal to 1 ppb; such a choice reflects the high NOx regime associated to biomass burning events. Two HCHO yields are computed: after one day of simulation (short-term), and after 2 months (final). The short-term yield is defined as

\[ Y_{st} = \frac{\text{HCHO produced after 1 day}}{C_0(\text{NMVOC})}, \] (1)

where \( C_0(\text{NMVOC}) \) is the initial concentration of the NMVOC. This yield represents the number of HCHO molecules generated by a given NMVOC one day after the injection time. The short-term yield defined in Eq. (1) provides better indication of the HCHO production that may be detected by the satellite instrument directly above biomass burning areas. The final yield is defined as

\[ Y_f = \frac{\text{HCHO produced}}{\Delta C(\text{NMVOC})}, \] (2)

where \( \Delta C(\text{NMVOC}) \) is the difference between the initial and the final NMVOC concentrations. Due to the importance of both short-term and final yields in the correct representation of the HCHO production by our chemical mechanism, particular care has been taken to ensure that the IMAGESv2 calculated HCHO yields are as close as possible to the MCM yields, as is evident from Table 1.

Compounds with the highest short-term HCHO yield are ethene (1.38 mol/mol), propene (1.78 mol/mol), and 2,3- butanedione (2 mol/mol). Strongly emitted compounds like acetic acid and methanol with lifetimes of several days have very small 1-day yields. For relatively short-lived species like ethene, glycolaldehyde, propene, acetaldehyde, and isoprene, more than 80% of the final yield is reached within the first day in the box model simulations.
Table 1: Photochemical production of HCHO from pyrogenic NMVOCs. The emission estimates are 10-year averages based on GFEDv2 and the MEGAN-ECMWF inventory. Short-term and final yields are obtained from box model simulations.

<table>
<thead>
<tr>
<th>NMVOC Emission (Tg/yr)</th>
<th>MCM HCHO Yield (mol/mol)</th>
<th>IMAGESv2 HCHO Yield (mol/mol)</th>
<th>MCM HCHO Production (Tg/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sh.-term</td>
<td>Final</td>
<td>Sh.-term</td>
</tr>
<tr>
<td>Acetic acid</td>
<td>17.1</td>
<td>0.12</td>
<td>1</td>
</tr>
<tr>
<td>Methanol</td>
<td>8.8</td>
<td>0.14</td>
<td>1</td>
</tr>
<tr>
<td>Ethene</td>
<td>6.3</td>
<td>1.38</td>
<td>1.76</td>
</tr>
<tr>
<td>Glyoxal</td>
<td>6.0</td>
<td>0.06</td>
<td>0.06</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>5.0</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Glycolaldehyde</td>
<td>4.8</td>
<td>0.80</td>
<td>0.87</td>
</tr>
<tr>
<td>Methylglyoxal</td>
<td>3.9</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>2,3-butanedine</td>
<td>3.7</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Acetaldehyde</td>
<td>3.6</td>
<td>0.94</td>
<td></td>
</tr>
<tr>
<td>Propene</td>
<td>3.5</td>
<td>1.78</td>
<td>1.92</td>
</tr>
<tr>
<td>Ethane</td>
<td>3.2</td>
<td>0.03</td>
<td>0.99</td>
</tr>
<tr>
<td>Acetone</td>
<td>3.0</td>
<td>0.11</td>
<td>2</td>
</tr>
<tr>
<td>Propane</td>
<td>1.9</td>
<td>0.03</td>
<td>1.65</td>
</tr>
<tr>
<td>2-butanone</td>
<td>1.7</td>
<td>0.53</td>
<td>2.15</td>
</tr>
<tr>
<td>Benzene</td>
<td>1.7</td>
<td>0.05</td>
<td>0.31</td>
</tr>
<tr>
<td>Toluene</td>
<td>1.0</td>
<td>0.62</td>
<td>1.34</td>
</tr>
<tr>
<td>Others</td>
<td>20.8</td>
<td>5.04</td>
<td></td>
</tr>
<tr>
<td>Total pyrogenic</td>
<td>96</td>
<td>35</td>
<td></td>
</tr>
</tbody>
</table>

4.1.3.2 HCHO yields from the oxidation of isoprene

The LIM1 isoprene oxidation mechanism (Peeters et al. 2014) is used in the current version of the model. This mechanism builds upon the previous version LIM0 (Peeters et al. 2009; Peeters and Müller, 2010; Stavrakou et al. 2010) and accounts for OH recycling in the oxidation of isoprene. To calculate the HCHO yields using LIM1, we conduct box model simulations starting at 9:00 am for a mid-latitude location in summer. Concentrations of NOx are kept constant throughout the simulation at very low NOx (40 ppt), low NOx (0.1 ppb), and high NOx conditions (1 ppb). The initial ozone concentration is set to 35 ppb. The HCHO yield from isoprene after one day of simulation is equal to 1.13, 1.54, and 2.20 mol/mol, and the final yields calculated after 2 months of simulation are equal to 1.78, 1.91, and 2.39 mol/mol, in very low, low, and high NOx regimes, respectively.

4.1.3.3 HCHO yields from the oxidation of anthropogenic NMVOCs

The MCM degradation mechanism for 32 anthropogenic NMVOCs is used in order to determine the HCHO yields from the NMVOCs oxidation under high (1 ppbv NO2) NOx conditions. Based on these yields and the species reactivity against OH oxidation, we derive the mean yield and mean reactivity for a lumped species OTHC (other hydrocarbons) which is implemented in IMAGESv2 (Table 2). In this way, OTHC is representative of the mixture of anthropogenic VOC compounds not explicitly included in the model. More details are given in Stavrakou et al. (in preparation).
Table 2: Anthropogenic VOCs and the corresponding HCHO yields (mol/mol) calculated in a high NOx environment.

<table>
<thead>
<tr>
<th>Species</th>
<th>Yield</th>
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<tbody>
<tr>
<td>butane</td>
<td>2.03</td>
</tr>
<tr>
<td>2-methyl butane</td>
<td>2.81</td>
</tr>
<tr>
<td>pentane</td>
<td>2.10</td>
</tr>
<tr>
<td>hexane</td>
<td>2.80</td>
</tr>
<tr>
<td>2-methyl propane</td>
<td>2.71</td>
</tr>
<tr>
<td>heptane</td>
<td>2.95</td>
</tr>
<tr>
<td>1,2,4-trimethyl benzene</td>
<td>3.00</td>
</tr>
<tr>
<td>ethylbenzene</td>
<td>1.11</td>
</tr>
<tr>
<td>octane</td>
<td>2.98</td>
</tr>
<tr>
<td>decane</td>
<td>3.83</td>
</tr>
<tr>
<td>2-methylpropene</td>
<td>2.89</td>
</tr>
</tbody>
</table>

4.1.4 Error analysis

Model errors might lead to potentially significant uncertainties on the derived fluxes which are, however, very difficult to estimate. To address this issue, sensitivity tests will be carried out to assess the influence of various parameters on the inferred fluxes. First, we will carry out inversions based on either GOME-2 or OMI HCHO columns, and assess the consistency between the derived emission fluxes. This will allow to investigate the robustness of the derived emissions on the choice of the satellite dataset. Furthermore, sensitivity tests where the error on the a priori emission parameters (biogenic, biomass burning) are either halved or doubled will be conducted. Another sensitivity experiment will consist in addressing the dependency of the results on the cloud filter assumed for the satellite data. For that, HCHO data with cloud cover lower than 20% will be used in the inversion, instead of 40% in the standard case study. Moreover, we have incorporated in IMAGESv2 a number of new developments in the isoprene degradation chemistry (Peeters and Müller, 2010; Stavrakou et al., 2010) which leads to HCHO formation. We will investigate the impact of these updates on the derived NMVOC flux strengths, especially over pristine forest regions, but also over mid-latitude regions in summertime. Further sensitivity studies could be conducted in order to improve the characterization of the uncertainties on the satellite-based emissions.
4.1.5 References


4.2 Regional Emission Estimates

4.2.1 Input data

The tropospheric columns of NO$_2$ and SO$_2$ of the satellite instruments OMI (on Aura) and GOME-2 (on Metop) are used. For both instruments, the tropospheric NO$_2$ columns are taken from the KNMI retrieval product (version 2), described by Boersma et al. (2011) and available through the TEMIS portal (http://www.temis.nl). SO$_2$ data is obtained from NASA/Goddard Space Flight Center (http://so2.gsfc.nasa.gov/).

4.2.2 Algorithm description

Below, we derive the algorithm for NO$_x$ estimation estimates, but it can be applied to other short-lived trace gases as SO$_2$ and CH$_2$O. Because of the rapid cycling between NO and NO$_2$, and the shift towards NO$_2$ during night time due to lack of photolysis, our analysis is based on the bookkeeping of NO$_x$ rather than NO$_2$, which is the observed quantity. We consider a time interval $t=[0,T]$ between two data assimilation moments, in our case the 24 h period between two overpasses of the satellite instrument. An schematic overview of the algorithm is depicted in Figure 3.

At the core of the algorithm is the chemical transport model (CTM) which calculates the concentration fields of NO and NO$_2$ at $t=T$ from the initial fields at $t=0$, given the meteorological conditions and a certain emission database. After the model run, however, there is no information available of the sensitivity of the final NO$_x$ column concentration on the emission field; information needed by the Kalman filter for the emission inversion. Instead, we will derive a simplified 2D transport equation which approximates this relation analytically. It uses trajectory analysis to transport NO$_x$ columns over the model domain. The aging of the NO$_x$ column is described by an effective lifetime, chosen in such way that it minimizes the calculated concentrations and the simulated concentrations by the CTM at $t=T$. The concentration fields from the CTM are also used to construct the forecasted tropospheric NO$_2$ column concentration, by interpolation towards the satellite footprint, extension with a
climatological column to the tropopause, and application of the averaging kernel from the satellite retrieval method.

**A priori emission inventory**

For the Chinese region, we use the recent INTEX-B emission inventory by Zhang et al. (2009, and private communication). It covers Asia on a 0.2°×0.2° resolution, containing the yearly totals of SO₂, NOₓ, CO, VOC, PM₁₀, PM₂.₅, BC, and OC by four sectors (power, industry, residential, and transportation) for the year 2006. The NOₓ emission totals are interpolated to the model grid and distributes over three chemical components: NO (90% of the NO equivalent mass of NOₓ), NO₂ (9.2%), and HONO (0.8%). The emissions are disaggregated to hourly values using sector-specific weekly and diurnal factors; no monthly cycle is postulated.

In analogy, the REAS emission inventory is used for the Indian region, and the MACCity emission inventory for the South African region.

**Chemical transport model**

The CHIMERE multi-scale model (Schmidt et al., 2001; Bessagnet et al., 2004) is primarily designed to produce daily forecasts of ozone, aerosols and other pollutants and make long-term simulations for emission control scenarios. In the presented configuration, CHIMERE has been implemented over East Asia (18°N to 50°N and 102°E to 132°E), simulating the atmosphere in 8 layers up to 500 hPa, with a horizontal resolution of 0.25°×0.25°. The meteorological data is taken from the deterministic forecast of the European Centre for Medium-Range Weather Forecasts (ECMWF), which is given on 91 atmospheric layers for a horizontal resolution of approximately 25×25 km². The boundary conditions for the domain border and top are taken from monthly climatologies (no nested run is performed). To reduce the effect of boundary values on the region of interest, the domain boundaries have been chosen over relatively unpopulated areas.

In Europe, CHIMERE has been extensively intercompared to other urban air quality models (e.g. Vautard et al., 2006) and evaluated against ground-based measurements and satellite data (e.g. Blond et al., 2007). For China, validation results can be found in Mijling et al., 2009.

**Derivation of the simplified 2D transport equation**

Let e, e(0), and e(T) be two-dimensional fields, representing the time-averaged emission, the NOₓ column concentration at t=0, and at t=T, respectively. They are written as vectors in an n-dimensional space, where n represents the total number of grid cells in the model domain. In our case, the dimension of state space n is 121×129=15,609.

For a certain grid cell at t=T, the NOₓ column is composed of an aged column from t=0 which has been transported into this grid cell, and a column of NOₓ which has been emitted during [0,T], aged, and transported into this grid cell. This can be written by the matrix equation:
\[ c(T) = Ge(0) + He \]  \hspace{1cm} (1)

Matrix \( G \) describes the advection and decay of the initial concentration \( c(0) \) over the model grid. Matrix \( H \) describes how the concentrations at \( t=T \) change if the emissions in time interval \( T \) change, i.e. its matrix elements represent the sensitivities of the \( \text{NO}_x \) column concentrations to the \( \text{NO}_x \) emission at the model grid:

\[ H_{ij} = \frac{\partial c_i}{\partial e_j} \]  \hspace{1cm} (2)

Both \( G \) and \( H \) have size \( n \times n \), but are sparse if transport to other grid cells covers only a small fraction of the model domain, which is true for short-lived species such as \( \text{NO}_x \) (~8h) and typical wind speeds (~24 km/h at 4 Beaufort) in our mesoscopic domain.

The first term in equation (1) describes the advection and decay of the initial concentration over the model grid. The concentration of \( \text{NO}_x \) particles in a certain grid cell column \( i \) at \( t=T \) can then be written as

\[ c_i^G(T) = \sum_j e^{-k_j T} \frac{a_j}{a_i} \Omega_{ij}(T)c_j(0) \quad \text{with} \quad k_j = 1/\tau_j \]  \hspace{1cm} (3)

in which the transport kernel \( \Omega_{ij}(T) \) represents the fraction of the area of cell \( j \) which is transported to cell \( i \) during time interval \( T \). To ensure mass conservation, the equation is scaled with the area \( a \) of the corresponding grid cells. For a regular grid over latitude and longitude \( a \) will depend on latitude \( \phi \): \( a/a_i = \cos(\phi_j)/\cos(\phi_i) \). The number of \( \text{NO}_x \) particles in the plume will decay exponentially because of aging, which is described by an effective lifetime \( \tau_j \) (or the reciprocal lifetime \( k_j \)) from its source location.

The second term in equation (1) represents all “fresh” \( \text{NO}_x \) which has been emitted at a certain moment in the time interval and subsequently transported during the remaining time. Analogous to equation (3), and integrating over all time dependent emission contributions \( e(t) \) during \( T \), we derive the expression:

\[ c_i^H(T) = \sum_j \frac{a_j}{a_i} \int_0^T e^{-k_j \cdot \Omega_{ij}(t)} e_j(T-t)dt \]  \hspace{1cm} (4)

The inversion only adjusts time-averaged emissions; the diurnal and weekly cycle of the emissions are described by an emission modulation function \( f(t) \), which is copied from the chemistry transport model:

\[ e_j(t) = f_j(t) e_j, \quad \left\{ f(t) \right\}_{\text{1 week}} = 1 \]  \hspace{1cm} (5)

Substituted in equation (4) this results in:

\[ c_i^H(T) = \sum_j \frac{a_j}{a_i} \left( \int_0^T e^{-k_j \cdot \Omega_{ij}(t)} f_j(T-t)dt \right) e_j \]  \hspace{1cm} (6)
Relating equation (4) and (6) to equation (1) gives us expressions for the elements of matrix $G$ and $H$ in terms of the (unknown) reciprocal lifetimes $k_j$ and the (known) transport kernel $\Omega(t)$:

$$G_y(k_j) = \frac{a_j}{a_i} e^{-k_j \tau} \Omega_y(T)$$  \hspace{1cm} (7)$$

$$H_y(k_j) = \frac{a_j}{a_i} \int_0^\tau e^{-k_j \tau} \Omega_y(t) f_j(T - t) dt$$  \hspace{1cm} (8)$$

We will call matrix equation (1) with matrix elements defined in (7) and (8) the simplified 2D transport equation. Because it does not contain boundary conditions, it does not account for NOx concentrations which are transported from the outside into the model domain. This will affect sensitivity relations $\partial c_i / \partial e_j$ (and consequently the assimilation results) close to the domain border. We suppress this boundary effect by choosing our domain borders (where possible) in remote areas with low emissions.

Equation (8) describes the sensitivity of the NOx concentration in cell $i$ on the NOx emission in cell $j$. However, we are interested in the sensitivity of the NO2 concentration on the NOx emission:

$$H^*_y = \frac{\partial c_{i,NO2}}{\partial e_j}$$  \hspace{1cm} (9)$$

From the CTM simulation we obtain the NO2/NOx ratio $\gamma_i$ for all cells $i$ at the moment of assimilation $T$:

$$c_{i,NO2}(T) = \gamma_i c_{i,NOx}(T) \hspace{1cm}, \hspace{0.5cm} 0 \leq \gamma_i \leq 1$$  \hspace{1cm} (10)$$

From this and equation (8) we get the sensitivities for NO2 column concentrations to NOx emissions, both at the model grid:

$$H^*_y(k_j) = \gamma_i H_y = \gamma_i \frac{a_j}{a_i} \int_0^\tau e^{-k_j \tau} \Omega_y(t) f_j(T - t) dt$$  \hspace{1cm} (11)$$

The transport kernel

Driving force behind the transport of NOx is the time and space dependent wind field $\mathbf{u}(x,y,z,t)$. In the simplified 2D transport equation the transport is described by matrix function $\Omega(t)$, whose column $j$ can be interpreted as the advection of the tropospheric column of NOx of grid cell $j$ over the model grid during the time interval $[T-t,T]$. We discretize $\Omega(t)$ at $n_T$ instants $t_n$, equally distributed over the assimilation time interval:
\[ \Delta t = T / n_T , \quad t_n = \left( n - \frac{1}{2} \right) \Delta t , \quad n = 1, \ldots, n_T \] (12)

If \( n_T \) is sufficiently large such that the transport distance is small during \( \Delta t \) (at wind speed \( v \) and grid cell size \( \Delta x \) this implies \( \Delta t << \Delta x / v \) must hold), the integral in equation (8) can be approximated by a summation over sparse matrix operations

\[ H_{ij}(k_j) \approx \frac{a_j}{a_i} \sum_{n=1}^{n_T} e^{-k_j \cdot \Delta \Omega_{ij}} f_j(T - t_n) \Delta t \] (13)

The elements of \( \Omega_{ij} \) are calculated for each \( t_n \) by trajectory analysis, which must take the divergence of the wind field and the height dependence of wind and NOx concentration into account. In remote areas, for example, the bulk of the NOx tends to be located higher up in the troposphere (mainly because of uplifting and washing out of the NOx in the lower troposphere) where wind flows generally faster. A practical method for fast calculation is by dividing the source grid cell \( j \) in a large amount of sub grid cells \( n_S \) and start the trajectory for each sub cell at a unique height, distributed according to the particle density in the NOx column. Each sub cell is transported along the time dependent, two-dimensional wind field vertically interpolated to this height. Then \( \Omega_{ij}(t) \) can be found by counting the number of sub cells which originate in cell \( j \) at time \( T - t \) and are within cell \( i \) at time \( T \).

The effective lifetime

The variation in lifetime, however, is too large to simply assume a fixed value for \( \tau \). The lifetime of NOx can vary between several hours and several days, depending on factors such as temperature, sunlight, precipitation, altitude, and presence of other chemical species and aerosols. This dependence is too intricate to make a straightforward parameterization of \( \tau \) which could be used in look-up tables. Instead, we use the results of the forward chemical transport model run to retrieve information on the NOx lifetime.

The matrix elements of \( G \) and \( H \) in equations (7) and (8) depend on the unknown reciprocal lifetimes \( k \) associated with the grid cells. The residue \( r \) is defined as the difference between the column concentrations \( c^{\text{CTM}} \) calculated with the CTM and the concentrations calculated with the simplified 2D transport equation, at \( t = T \):

\[ r(k) = c^{\text{CTM}}(T) - \left( G(k)c^{\text{CTM}}(0) + H(k)e \right) \] (14)

The effective lifetime field is found by minimizing the residue for \( k \) numerically. The exponential dependence in \( k \) of the matrix elements, and the reciprocal dependence of \( \tau \) of \( k \), makes the residue function relatively insensitive for variations in both low and high values of \( \tau \). Unrealistic high or low lifetime values are corrected by constraining the lifetime \( \tau \) between 2 h and 48 h. Far from emission sources, the lifetime can only be determined from the decay of the small background field, which contributes very weakly to the residue function. A good a priori lifetime field is therefore essential, and is taken from the results of the previous day, assuming that day-to-day variations are small.
Comparing model simulations with satellite observations

For NO\textsubscript{2}, not all satellite retrievals are included in the data assimilation. Retrievals at cloudy conditions (cloud fractions larger than 20\%) are filtered out, to reduce the influence of the modelled NO\textsubscript{2} column below the clouds in the retrieval. For bright surfaces such as snow (surface albedo larger than 20\%) the cloud fraction from the cloud algorithm becomes uncertain, hence these retrievals are also discarded. Retrievals with clouds below 800 hPa are filtered out because the possible intersection of the cloud with the NO\textsubscript{x} bulk makes the retrieval too sensitive for the exact cloud height. For OMI, the large outer 4 pixels at either side of the swath are filtered out, as are pixels affected by the row anomalies (which appear since June 2007). For GOME-2 we discard the backscan pixels (which are too large), and retrievals in the zoom-mode of the instrument (which are of unknown quality).

The CHIMERE model calculates daily tropospheric columns up to 500 hPa. In order to extend the NO\textsubscript{2} profiles from the model ceiling to the tropopause, we add a climatological partial column for this part of the free troposphere. This climatology was compiled from a 2003–2008 run of the global chemistry transport model TM5, described in Huijnen et al. (2010), at a 2°×3° resolution and 34 atmospheric layers. Although in populated areas the added free tropospheric column contributes only a few percent to the total tropospheric column, it can account for up to 50\% in remote areas, where the tropospheric NO\textsubscript{2} column is small.

We construct a representative vertical NO\textsubscript{2} profile \( p_i \) for the footprint area by taking a weighted average of the modeled vertical profiles \( p_c^j \) in contributing grid cells, taking weight factor \( W_{ij} \) from the surface fraction of grid cell \( j \) which is covering footprint \( i \) :

\[
 p_i = \sum_j W_{ij} p_c^j = W p^c, \quad \text{with } W_{ij} \text{ normalized } \sum_j W_{ij} = 1
\]  

Afterwards, the interpolated profile \( p_i \) is rebinned to the model layers used in the tropospheric NO\textsubscript{2} retrieval algorithm, so the averaging kernel \( A_i \) can be applied to get the modeled, corrected NO\textsubscript{2} column at satellite footprint \( i \) (which is the observable quantity):

\[
 y_i = A_i p_i
\]  

For the inversion we will also need the Jacobian of the model, i.e. the matrix \( H \) which represents the linearization of the model around a certain emission field \( e \):

\[
 \Delta y = H \Delta e
\]  

\( H \) describes the sensitivity of the modeled observations \( y \) (in observation space) to changes in model emissions \( e \) (in state space). We want to find an expression of \( H \) in terms of the model sensitivities we found in equation (11). The gridded concentrations are interpolated to observation footprints by applying the interpolation matrix \( W \) from equation (15):

\[
 \Delta \tilde{y} = WH \Delta e
\]  

in which \( \Delta \tilde{y} \) represents the change of the total tropospheric NO\textsubscript{2} columns in observation space, simulated by our model. Note that there is no information on the vertical profiles,
which complicates a direct application of the averaging kernels. Therefore we assume that an emission change $\Delta e$ does change the total column value $\widetilde{y}_i$ for modeled observation $i$, but does not change its vertical profile shape, so we can write the change in averaged, modeled observation in terms of the change of modeled observations:

$$\Delta y_i = \alpha_i \Delta \widetilde{y}_i, \quad \alpha_i = \left( \frac{A_i}{y'_i} \right)$$

(19)

Combining equations (18) and (19), we get the expression for the Jacobian, in terms of the model sensitivities, the interpolation matrix, and the averaging kernels:

$$\Delta y_i = \alpha_i \sum_j w_{ij} H_{jk} \Delta e_k \Rightarrow H = \text{diag}(\alpha)W \mathbf{H}^\top$$

(20)

in which $\mathbf{H}^*$ is the $(n \times n)$ model sensitivity matrix described by equation (11) for NO$_2$ columns to NO$_x$ emissions on the model grid, and $\mathbf{H}$ is the $(m \times n)$ sensitivity matrix for NO$_2$ column observations to gridded NO$_x$ emissions, to be used in the Kalman equations below.

**The Kalman filter for emission estimation**

The relation between column concentration difference vector $\Delta y$ and emission update vector $\Delta e$ can be written as $\Delta y = H \Delta e$. Note that solving $\Delta e$ from $\Delta y$ and $H$ is an underdetermined problem. Furthermore, the errors in $y$ are large, and they would propagate non-locally in the solution, causing strong fluctuations in the emissions if this assimilation scheme is applied iteratively. To deal with these issues we use the Kalman filter, which calculates for every assimilation step the most probable emission field and its covariance, taking into account the errors in the modeled emissions and representation, and the errors in the observed concentrations. Due to the non-linearity of the problem the extended Kalman filter is used, which linearizes about the current mean and covariance:

- **State vector forecast**
  $$\mathbf{e}^f(t_{i+1}) = M_i [\mathbf{e}^a(t_i)]$$
  (K1)

- **Error covariance forecast**
  $$\mathbf{P}^f(t_{i+1}) = M_i \mathbf{P}^a(t_i) M_i^T + \mathbf{Q}(t_i)$$
  (K2)

- **Kalman gain matrix**
  $$\mathbf{K}_i = \mathbf{P}^f(t_i) H_i^T [H_i \mathbf{P}^f(t_i) H_i^T + \mathbf{R}_i]^{-1}$$
  (KG)

- **State vector analysis**
  $$\mathbf{e}^a(t_{i}) = \mathbf{e}^f(t_{i}) + \mathbf{K}_i (y_i - H_i [\mathbf{e}^f(t_{i})])$$
  (K3)

- **Error covariance analysis**
  $$\mathbf{P}^a(t_{i}) = (I - \mathbf{K}_i H_i) \mathbf{P}^f(t_{i})$$
  (K4)

The interpretation of the quantities is as follows:

- $\mathbf{e}^a, \mathbf{e}^f$ analysis and forecast of the NO$_x$ emissions.
- $\mathbf{P}^f, \mathbf{P}^a$ error covariance matrices $(n \times n)$ of the forecasted emissions $\mathbf{e}^f$ and the analysis of the emissions $\mathbf{e}^a$.
- $M$ model describing temporal evolution of the emissions. $M$ applied on the true state is assumed to introduce no bias: $\mathbf{e}^f(t_{i+1}) = M_i [\mathbf{e}^f(t_{i})] + \mathbf{e}_i$, in which the noise is normally distributed around 0 with covariance $\mathbf{Q}$.
- $\mathbf{Q}$ covariance matrix $(n \times n)$ of the modeled emissions (see Section 0).
$M$ $(n \times n)$ matrix representation of emission model $M$.

$y$ observations of tropospheric NO$_2$ columns.

$H$ observation operator which relates the emissions of NO$_x$ in the model grid to the observable tropospheric column concentrations of NO$_2$. $H$ applied to the true state is assumed to introduce no bias: $y_0^i = H[e(t_i)] + \varepsilon_i$, in which the noise is normally distributed around 0 with covariance $R$.

$R$ $(m \times m)$ matrix describing the covariances of the observation operator $H$ (see Section 0).

$H$ $(m \times n)$ Jacobian of the observation model $H$, linearized around state $e=e^a$:

$y^f = y^a + H[e^f-e^a]$, describing how the tropospheric NO$_2$ column of observation $i$ changes when the NO$_x$ emission $\varepsilon$ in grid cell $j$ is changed.

### Covariance of the observation minus forecast

The covariance of the observation minus forecast of the column concentration consists of three independent components:

$$\Sigma_{Omf} = \Sigma_{obs} + \Sigma_{repr} + H^T H = R + H^T H$$

(21)

The observation error of the tropospheric NO$_2$ column is composed of errors by the measurement noise of the satellite instrument and the spectral fitting, errors related to the separation of the troposphere and the stratosphere, and errors due to retrieval method parameters, such as clouds, surface albedo and a priori profile shape (Boersma et al., 2004). The second part describes representation error originating from an inaccurate CTM (due to errors in e.g. meteorology or chemistry scheme), and errors introduced by adding a climatological free tropospheric column, and interpolating grid values to the satellite footprint. Together with the observation errors they are contained in covariance matrix $R$. The last part of the OmF error describes how errors in the emission estimation propagate into the simulated column concentrations. The sensitivity matrix $H$ is assumed to be exactly known; the error made by the approximating $H$ with the simplified 2D transport equation is added to $R$.

It is the balance between $R$ and $H^T H$ which determines how much information from the observed concentration difference is used to update the emission estimates. We work out a practical method to estimate the covariance matrix $R$. By neglecting spatial correlations the OmF error $\sigma_{omf}$ for observation $i$ can be written, analogous to equation (21) as

$$\sigma_{omf,i}^2 = \sigma_{obs,i}^2 + \sigma_{repr,i}^2 + \sigma_{prem,i}^2 = \sigma_{R,i}^2 + \sigma_{prem,i}^2$$

(22)

in which $\sigma_{prem,i}^2$ is the propagated emission variance of matrix $H^T H$. The observation error $\sigma_{obs}$ is known from the satellite product: typically they have a dominating absolute error at low values (around 0.5 $10^{15}$ molecules/cm$^2$), and a dominating relative error at high values (around 30–45%). The representation error $\sigma_{repr}$ is unknown, but is assumed to be relative to the simulated tropospheric column concentration $y$ with a fixed $\varepsilon_{rel}$. 

\[ \sigma_{\text{repr},i} = \varepsilon_{\text{rel},i} y_i \]  

(23)

For each assimilation a large quantity of observations and forecasts are available, enabling the calculation of \( \sigma_{\text{repr}} \) from OmF statistics, using the reduced \( \chi^2 \) criterion:

\[ \chi^2_{\text{red}} = \frac{1}{m} \sum_i \left( \frac{y_{\text{obs},i} - y_i}{\sigma_{\text{OmF},i}} \right)^2 \]  

(24)

We calculate \( \varepsilon_{\text{rel}} \) such that \( \chi^2_{\text{red}} = 1 \), meaning that the variation in OmF is well described by its error \( \sigma_{\text{OmF}} \): the distribution of \( (y_{\text{obs},i} - y_i)/\sigma_{\text{OmF}} \) will be Gaussian around 0 with standard deviation 1. By substituting (22) and (23) in (24) we find the equation:

\[ f(\varepsilon_{\text{rel}}) = \sum_{i=1}^{m} \frac{(y_{\text{obs},i} - y_i)^2}{\varepsilon_{\text{rel},i}^2 y_i^2 + \sigma_{\text{obs},i}^2 + \sigma_{\text{prem},i}^2} - m = 0 \]  

(25)

We can find the root of \( f \) numerically using its derivative to \( \varepsilon_{\text{rel}} \) in Newton’s method (e.g. for OMI observations in May 2008, \( \varepsilon_{\text{rel}} = 33\% \)). With the daily estimated value of \( \varepsilon_{\text{rel}} \) we calculate \( \sigma_{R,i} \) according to (22). From these errors we construct the covariances by decomposing the error covariance matrix as

\[ R = \text{diag}(\sigma^2) C \text{diag}(\sigma^2) \]  

(26)

in which \( C \) is a correlation matrix which elements only depend on the distance between two observation footprints: \( C_{ij} = g(r_{ij}) \). We model \( g \) exponentially dependent on distance:

\[ g(r_{ij}) = \exp(-r_{ij} / L) \]  

(27)

in which \( L \) is the correlation length. Best inversion results are obtained by taking \( L \) small with respect to the footprint size (we will use \( L = 10 \) km); in this case the improved condition of the matrix which is inverted in (KG) suppresses spatially oscillating solutions. Correlations between footprints at a distance larger than 6\( L \) are considered insignificantly: corresponding matrix elements \( C_{ij} \) are set to zero, resulting in a sparse covariance matrix \( R \).

### Emission covariances and inversion behavior

For the Kalman filter approach we need to assess the emission model \( M \), its error covariance \( Q \). For these matrices it is important to find realistic estimates, which will optimize the assimilation for both convergence speed and noise reduction.

Anthropogenic emission trends, if present, are usually very gradual, justifying a persistent emission model, stating that tomorrow’s emissions are equal to today’s emissions. This implies that the emission model reduces to the identity matrix: \( M = I \). Note that our algorithm adjusts averaged emission totals; the diurnal and weekly cycle is modulated already by the CTM, see equation (5).

To assess the influence of the emission covariance \( Q \) on the inversion behavior we analyze
the Kalman equations for the simple case where the concentration \( y \) only depends on one emission source \( x \) with a constant sensitivity factor \( h \). The Kalman filter reduces to the following scalar equations, and matrix \( Q \) becomes a scalar quantity \( q \), dictating how much the error of the emission increases between two assimilation moments. We find an expression for the evolution of the error analysis \( \sigma_i^a \) in terms of its predecessor \( \sigma_{i-1}^a \):

\[
\sigma_i^2 = \frac{\sigma_{R,j}^2}{h^2(\sigma_{i-1}^2 + q^2)} + \sigma_{R,j}^2
\]

(28)

We can see with this formula that the limit value of the assimilation error \( \sigma^a \) depends on the sensitivity \( h \) (a higher \( h \) results in faster convergence to a lower \( \sigma^a \)), the observation and representation error \( \sigma_R \) (a lower \( \sigma_R \) results in a lower \( \sigma^a \)), and emission error increment \( q \). \( q \) is the only unknown parameter and its value will affect the noise and convergence in the assimilation. A high emission uncertainty \( q \) results in a large noise on the assimilated emission, and a low uncertainty \( q \) leads to slow convergence.

We model \( q \) with a dominating absolute error \( \varepsilon_{abs} \) at low emissions, shifting to a dominating relative error \( \varepsilon_{rel} \) at high emissions:

\[
q(x) = \varepsilon_{abs} \exp(-\varepsilon_{rel}x / \varepsilon_{abs}) + \varepsilon_{rel} x
\]

(29)

Note that the absolute error component is essential to be able to pick up changing emissions in areas where zero emissions are defined. For our assimilation setup we construct matrix \( Q \) analogous to (26) and (27), modeling its errors according to (29) with \( \varepsilon_{abs}=0.02 \times 10^{15} \) molecules/cm\(^2\)/h and \( \varepsilon_{rel}=5\% \), and allowing for weak covariances with its nearest neighbors by taking the covariance length \( L=10 \) km. These values are a trade-off between convergence speed and noise reduction, without losing the possibility to pick up new emission sources.

The Kalman gain

After establishing the covariance matrices as described in the above sections, the Kalman equations can be solved numerically. Calculation time and storage space is reduced by making optimal use of the sparseness of the matrices. Point of concern is the inversion of the symmetric matrix \( A=HPH^T+R \) in the Kalman Gain (equation (KG)), which generally is ill-conditioned and contains a null-space. Note that \( A \) being symmetric and positive semi-definite implies that \( A \) has real, positive eigenvalues. The eigenvalues spectrum of matrix \( A \) consists of only a few large eigenvalues and many smaller ones, which makes solving the inverse of \( A \) very sensitive to noise. This sensitivity issue can be avoided by approximating \( A \) with a decomposition

\[
A \approx U \Lambda U^T \Rightarrow \Lambda^{-1} \approx U \Lambda^{-1} U^T
\]

(30)

in which \( \Lambda \) is a diagonal matrix consisting of the \( m_t \) largest eigenvalues of \( A \). Here we use the numerical software library ARPACK (Lehoucq et al., 1998) for a fast calculation of the largest eigenvalues and corresponding eigenvectors. We take \( m_t \) such that the sum of the largest eigenvalues account for 98% of the value of the trace of \( A \), with a practical maximum
of 1800 eigenvalues. As a result the condition number (the ratio of the largest eigenvector to the smallest eigenvector) is reduced to an order of 1000.

**Calculation of the analysis covariance**

Correlations in the emission analysis are introduced because different emission grid cells contribute to an observation in the satellite footprint. Although the covariances can be strongly non-local (e.g. when fast winds transport long living NO$_x$ over remote areas), normally the covarying emission area is localized within a certain distance of the grid cell. Therefore it is not necessary to calculate all $n \times n$ elements of the analysis covariance matrix $P^a$. Instead we calculate the diagonal elements $P^a_{ii} = \sigma_i^2$ using (K4). Off-diagonal, we only calculate the covariance for grid cells within a radius of 300 km. Only correlations larger than 0.01 are supposed to contribute significantly to the analysis:

$$|\rho_{ij}| = \left| \frac{P^a_{ij}}{\sigma_i \sigma_j} \right| > 0.01$$  \hspace{1cm} (31)

By neglecting all smaller correlations the analysis covariance matrix becomes a sparse matrix.

**Emission inventory update**

After calculating new emissions we have to make certain assumptions to use this data to update the emission database. At this time we only adjust the NO$_x$ emissions, neglecting the possibility that a change in anthropogenic NO$_x$ is related to a change in other anthropogenic emissions such as particulate matter and CO. Furthermore, we assume that the emission change is due to anthropogenic sources. Since we do not know how the new NO$_x$ emissions can be attributed to the different sectors (e.g. power, industry, residential, and transportation in the INTEX-B inventory for the Chinese region), we scale emissions in all sectors by ratio, assuming that the introduced error (through different sectorial diurnal and weekly emission cycles) is small. For new emission sources, where no sector information is known, the new NO$_x$ emissions are distributed evenly over all sectors. Finally, information about the injection height of the new emission cannot be inferred with the DESCO algorithm. Instead, emissions at all heights will be scaled by ratio.

**4.2.3 Error analysis**

The Kalman filter keeps track of the error estimates of each grid cell emission. The error of the emission analysis depends on the sample frequency of the downwind concentration plume, the average sensitivity of the emission to the observed concentration, and the error growth of the emissions between assimilation moments. In Figure 4 the emission analysis errors of the entire domain are plotted against the emissions, and are modelled according to equation (29). For OMI data assimilation, we find a dominating absolute error of $0.38 \times 10^{15}$ molecules/cm$^2$ at low emissions, and a dominating relative error of 47% at high emissions. Data assimilation with GOME-2 results in larger emission errors due to the smaller amount of
observations: $0.48 \times 10^{15}$ molecules/cm$^2$ at low emissions, and 74% at high emissions. Note that when the emissions converge to their true state the analysis errors become overestimated due to an overestimation of the emission model covariance $Q$. However, inflating the emission forecast covariance by a significant $Q$ at every assimilation cycle is necessary to avoid becoming insensitive to emission changes.

Calculating the emission for an area larger than a grid cell reduces the associated error considerably. For example, for the individual Beijing grid cells in the OMI time series of May-December 2008 the mean relative emission error is 58%. By taking an average over 4 grid cells, the mean error drops to 23%. By taking the negative covariances between the grid cells into account the emission analysis error drops further to 20%. For the GOME-2 time series, the errors are 91%, 47%, and 38%, respectively. The effect of the negative covariances is here stronger for GOME-2 due to the larger footprint size.

Figure 2: Scatter plot for emission analysis against its error, for all emissions in the domain at 1 December, 2008. Left panel shows results for OMI, right panel for GOME-2.
4.3 Emission Inventories for European products

Establishing (long term) trends in pollutant emissions and concentrations is a key part of evaluating the impact of policies. Traditionally, concentrations of air pollutants are monitored using in-situ measurement networks (Tørseth et al., 2012), whereas emissions are estimated on annual basis within the convention for long range transport and air pollution (CLRTAP). Establishing trends based on monitoring networks is hampered by different equipment used at individual sites or countries, replacement of instruments, etc. (Cooper, Gao, Tarasick, Leblanc, & Sweeney, 2012; Sicard, Coddeville, & Galloo, 2009). Moreover, large areas in (south eastern) Europe are not covered by these networks. Similarly, the approaches and quality of emission reporting are variable among European countries (Pouliot et al., 2012). Hence, trend analyses based on satellite data may provide a valuable independent source of information to compliment traditional monitoring strategies (Bovensmann et al., 1999; Levelt et al., 2006).

To determine the trends in anthropogenic NOx across Europe two approaches are investigated. In the first approach, a fit model is applied to the timeseries of the bias between modelled and retrieved NO2 tropospheric column. It is assumed that the NO2 column change is representative for an emission change and its validity assumption has been tested by Schaap et al. (2013). In the second approach Ensemble Kalman Filter is applied to ingest the retrieved NO2 tropospheric column in the LOTOS-EUROS model. This approach allows for a better capture of the variability in emission strengths as they allow for the estimation of emission variability that changes sign or slope within the period of interest.

4.3.1 Input data

4.3.1.1 Meteorological data

The model is driven by 3-hourly meteorological data provided by European Centre for Medium-range Weather Forecast (ECMWF). These include 3D fields for wind direction, wind speed, temperature, humidity and density, substantiated by 2-d gridded fields of mixing layer height, precipitation rates, cloud cover and several boundary layer and surface variables. The vertical velocity field is calculated using the horizontal wind fields and the mass conservation law of incompressible fluids. Further, the water vapour concentration is calculated using the Claussius-Clapeyron relation. Rain is neglected when the 3-hour accumulated amount is less than 0.3 mm. Linear interpolation is used to derive the meteorological fields at the interval times between the update times.

4.3.1.2 Emissions

The major driver of the LOTOS-EUROS system is the anthropogenic emission data of VOC, SOx, NOx, NH3, CO, CH4 and PM. The anthropogenic emission were prescribed following the MACC emission database ([Kuenen et al., 2011]). The annual emission totals reported for 2005 have been converted to hourly emission estimates using time factors for the emissions strength variation over the months, days of the week and the hours of the day and scaled for all year between 2003-2007. To account for the occasional fire events, the MACC global fire assimilation system, [Kaiser et al., 2009], is used on a hourly basis. The biogenic NMVOC
emissions are calculated online following (Steinbrecher et al., 2009) and the sea salt emissions are parameterised following source formulations for coarse (Monahan et al., 1986) and fine (Mårtensson et al., 2003) aerosol modes.

4.3.1.3 Boundary conditions

The model is constrained by boundary conditions used from the global MACC reanalysis (Hollingsworth et al., 2008, Schere et al., 2012) based on a coupling of the ECMWF model [Flemming et al., 2009] to the chemical transport model MOZART [Emmons et al., 2010].

4.3.2 Algorithm description

4.3.2.1 LOTOS-EUROS

The LOTOS-EUROS model is an operational air quality/chemical transport model of intermediate complexity focused on modelling the lower part of the troposphere. Below a description is given of the model characteristics. In this study LOTOS-EUROS model (version 1.10.001), the domain spans from 35° to 70°Nord and -10° to 60° East with a grid resolution of 0.5° longitude x 0.25° latitude (approximately 25x25 km at mid latitude).

In the vertical the model has four layers up to 3.5 km above sea level: a fixed surface layer of 25 meter and three dynamic layers. The lowest dynamic layer is the mixing layer, followed by two equally thick reservoir layers up to the model top. The height of the mixing layer is part of the meteorological input data. The height of the reservoir layers has a minimum of 50 m. In some cases when the mixing layer extends near or above 3.5 km the top of the model exceeds the 3.5 km according to the abovementioned description. For output purposes the concentrations at measuring height (reference height is usually 3.6 m) are diagnosed by the constant flux approach which relates the dry deposition speed and the concentration of a pollutant.

The chemistry is parameterized following the TNO CBM-IV scheme [Schaap et al., 2008] and the aerosol chemistry is accounted for using the ISORROPIA parameterization (Fountoukis & Nenes, 2007). The transport is represented by advection in three dimension, vertical diffusion and entrainment. The dry deposition is parameterized following the resistance approach (Erisman et al., 1994, Wichink Kruit et al., 2012). The wet deposition process is represented by below cloud scavenging for gases (Schaap et al., 2004) and particles (Simpson et al., 2003)

4.3.2.2 Trends analysis: Remnant approach

For the trend analysis the satellite (OMI and GOME-2) observations are used in synergy with the LOTOS-EUROS model. A multi-year simulation is performed using a single and thus constant a-priory anthropogenic NOx emission database. We use the model to estimate the variability due to synoptic variability in weather systems, which it resolves quite well. Moreover, the model resolves part of the seasonal variability due to changing in chemistry and mixing. As we are using a fixed NOx emission database the bias between the observations and model results will systematically change as the real emission strength is changing. The changes in the bias can therefore provide fruitful information concerning
changes in NO$_2$ tropospheric columns arising from changes in the emission in the recent years.

In this study we apply a fit model (Weatherhead et al., 1998) to the time series of the bias between model and observation (hereafter called the remnant). In previous studies this fit model was used on NO$_2$ tropospheric columns using GOME and SCIAMACHY data at a global scale and over China (van der A et al., 2006, van der A et al., 2008). The fit model can be described by the following function:

$$Y_t = C + \frac{1}{12} B t + A \sin \left( \frac{\pi}{6} t + \alpha \right) + N_t$$

where $Y_t$ is the remnant at month $t$. The first two terms represent a linear trend with slope $B$ representing the annual change in NO$_2$. The third term describes the seasonal component of the annual cycle in de bias with amplitude $A$ and phase shift $\alpha$.

$N_t$ is the remainder which cannot be explained by the fit model. The autocorrelation, $\phi$, in the remainder affects the precision of the trend. The autocorrelation in the remainders was analysed and an average autocorrelation of 0.1 was found without an indication of a spatial pattern. Hence, the remainders are weakly correlated and the trend precision is determined using the average value. The linear trend $B$ is considered as statistically significant with a 95% confidence level when $|B/\sigma_B| > 2$, where $\sigma_B$ is the precision of the trend. $\sigma_B$ is defined as a function of the autocorrelation, the length of the dataset in months and the variance of the remainder, $\sigma_N$:

$$\sigma_B = \frac{\sigma_N}{n^{3/2}} \sqrt{\frac{1 + \phi}{1 - \phi}}$$

The fitting model (Weatherhead et al., 1998) is applied to each grid cell across the domain of interest, providing a spatial distribution of the fitting parameters.

Figure 5 shows an example of a measured timeseries and the fitted function. The monthly averaged NO$_2$ tropospheric column is plotted as a function of the month number starting in January 2005. The black dots represents the measurement while the red and blue lines represent respectively the linear decrease and seasonal component of the fitting result.
4.3.2.3 Trends analysis: Kalman filter approach

The trends analysis of the bias assumes that the NO2 column changes is representative for an emission change. For most of the larger countries the assumption is valid, however for smaller countries and in the case of a strong increase in specific sectors such as international shipping the trends in the columns may underestimate actual emission trends of a country. To account for these issues, the chemistry transport model can be used to a larger. In practice, LOTOS–EUROS is equipped with a data assimilation package (Barbu, Segers, Schaap, Heemink, & Builtjes, 2009; Curier et al., 2012), which is used to assimilate the OMI NO2 tropospheric column. To estimate the change in NOx emission from year to year, data-assimilation of OMI tropospheric NO2 measurements in the LOTOS-EUROS chemistry transport model will be performed. The variability in the NOx emission scaling factor from the EnKF should represent the inter-annual changes in the NOx emissions.

Assimilation system

A data assimilation system has been developed around the LOTOS-EUROS model based on the Kalman filter technique. A Kalman filter computes probability density functions (pdf's) of the true state, given 1) a transition model to propagate the state in time with associated uncertainties; and 2) observations with associated representation error. Starting from initial pdf, the filter first performs a forecast step propagating the pdf in time to the first moment that observations become available. Then, during the analysis step, the forecasted pdf is replaced by an analyzed version that takes into account the new information that has become
available. The Kalman filter is an example of a sequential assimilation, since forecast and analysis steps follow each other sequentially in time and use only information from the past.

The original linear Kalman Filter is closed, i.e. if the initial pdf, the model uncertainty, and the representation error are expressed as Gaussian (normal) random vectors, and if in addition the transition model is linear, then the computed pdf's for the true state are Gaussian too. The advantage of Gaussian random vectors is that the complete pdf is described by a mean state vector and a covariance matrix only. In practice, the linear Kalman filter cannot be implemented for large scale applications as the size of the state vector is usually very large (at least \(n=10^4\) elements) and storage of a covariance matrix becomes impossible since it requires \(n^2\) elements. In addition, the linear Kalman filter requires \(2n\) evaluations of the transition model, which is far too expensive for the type of models considered here. Moreover transition models are usually non-linear as chemical reactions are included.

A suitable alternative for the linear Kalman filter for air pollution models is the Ensemble Kalman filter (EnKF) (Evensen 1994). In this formulation, the pdf of the state is not expressed in terms of a mean and covariance only, but is described by an ensemble of model states. The spread between the ensemble members should describe the uncertainty in the value of the state and quantities as mean and covariance of the state are computed from the ensemble statistics. The transition model is therefore not restricted to linearity, which simplifies the actual formulation and allows for a transparent implementation. The number of required ensemble members depends on the complexity of the pdf to be described, which is usually determined by the non-linearity of the transition model and the complexity of the associated model uncertainty. In practice, an ensemble with 10-100 is acceptable to keep computations feasible, and the complexity of the problems is limited to a point that this number is reached.

In the EnKF formulation around the LOTOS-EUROS model used in this study, the basic time step between two analyses is one hour. At the end of each step, all observations that have become available are analyzed. The number of analyzed observations therefore depends on the overpass time of the satellites.

**Transition model**

The transition model from time (day) \(t[k]\) to \(t[k+1]\) is given by:

\[
\begin{align*}
    c[k+1] &= M(c[k],\lambda[k],z[k]) + \frac{o}{\sqrt{1 - \alpha^2 \sigma^2}} \\
    \lambda[k+1] &= M(x[k]) + \Gamma w[k]
\end{align*}
\]

Equation 1

The transition model consists of 2 parts:

- The vector \(c[k]\) contains the instantaneous model state at the end of the time step (here hourly). The model state consists of the concentrations of all components in each grid cell, as well as concentration depended fields such as the total aerosol water content. Operator \(M\) denotes the propagation of the concentrations by the LOTOS-EUROS model from hour to hour.
- The model is driven by uncertainty parameters \(\lambda[k]\), which are kept constant during the time step.
In the current application, uncertainty is defined for the emission input. Emissions from anthropogenic as well as natural emissions are considered uncertain, to enforce variation in the studies key species O3 and NO2 but also in HCHO are CO, where the later are strongly influenced by natural emissions. Every time the model simulates emissions, the actual emission of group p in grid cell ij is described by:

\[ s_{ij}^p[k] = \bar{s}_{ij}^p(1 + \lambda_{ij}^p[k]), \]

where \( \lambda \) denotes the nominal value. To account for emissions just outside the domain, the boundary conditions are changed similar to the adjacent emissions. The uncertainty \( \lambda \) is described as a colored noise process driven by an \( r \)-element uncorrelated white noise \( w[k] \) with zero mean and identity covariance; each element accounts for a different emission field. The standard deviation of the uncertainty is set to a value \( \sigma \) which has to be determined. To ensure that only realistic emissions are used, the values of \( \lambda \) are bounded into the interval \([-1,1]\), such that emissions are never non-negative or more than doubled. A temporal correlation is assumed with exponential decay; by this, the value of the emissions will change gradually from day to day, to ensure that emissions are lower or higher than the nominal value during a longer period. The driving white noise \( w[k] \) has only \( r \) elements (one for each group of uncertain emitted components), no spatial variation is initially included in the emission uncertainty. However, since the noise factors \( \lambda \) are available for each grid cell, spatial variations will be introduced during the analysis step.

The uncertainty specification has a number of undefined settings yet, which have to be defined prior to the assimilation experiments. These settings include the selection of the uncertain emissions (source categories, emitted species), the amplitude of the assumed uncertainties, and their temporal correlation. Appropriate values are those that can explain the observed difference between the model simulations and the observations, taking into account the representation error. These values will therefore be chosen once the (synthetic) observations are available.

**Ensemble formulation**

An ensemble of \( m \) state vectors forms the main data structure in the EnKF:

\[ \xi_j[k], \quad j = 1, \ldots, m \]

The spread in the ensemble should at any time represent the uncertainty about the value of the true state \( x[k] \); the true value exists but is unknown to the user, who could only provide an estimate of what are likely values. A mean and covariance for the unknown true state can be computed from the ensemble:

\[ \bar{x} = \frac{1}{m} \sum_{j=1}^{m} \xi_j \]

\[ P = \frac{1}{m-1} \sum_{j=1}^{m} (\xi_j - \bar{x})(\xi_j - \bar{x})^T \]

The ensemble members are initialized at day \( k=0 \) with 'analysed' filter states \( \xi_j^a[0] \), which are all the same and include model states \( c[0] \) resulting from a model run over the two weeks.
prior to the assimilation period, and uncertainty parameters $\lambda[0]$ drawn from random
generator with zero mean and standard deviation $\sigma$. In the forecast stage of the filter, each
ensemble member $\xi_j$ is propagated over the next time step using a white noise sample $w_j$
drawn from a random generator:

$$\xi_j[k] = M(\xi_j[k-1]) + \Gamma w_j[k-1]$$

**Observation representation**

At the end of each step, the (synthetic) observations that have become available during this
time step are collected in a vector $y[k]$. The observations can be simulated from a filter state
by an operator $H$; the operator extracts from the concentration array the value at the station
location, or the (partial) column over the footprint of a satellite pixel taking into account the
averaging kernel. The difference between the observations and the simulations from the
(unknown) true state is supposed to be described by:

$$y[k] - H[k]x[k] = v[k]$$  \hspace{1cm} \text{Equation 6}

where $v[k]$ is a random vector with zero mean covariance $R[k]$. These representation errors
(or residues) are supposed to be uncorrelated ($R$ is diagonal). The standard deviations that
define the diagonal have to be determined by examining the difference between the
simulations and the (synthetic) observations, taking into account the model uncertainty to
(see previous section on uncertainty model). A parameterization for the representation error
standard deviation could be that it is equal to a fraction of the observed value, with thresholds
for the minimum and maximum values.

For representation of satellite observations, also the size of the footprints with respect to the
model grid cell size should be taken into account. The spatial representation error that is
made by averaging concentrations over the footprint could be determined by comparing
model simulations at the highest resolution with fields obtained after spatial averaging. This
procedure has been applied to the LOTOS-EUROS simulations of formaldehyde of the nature
run; the results are shown in Figure 1.

![Figure 1: Variance of difference between high resolution simulations of formaldehyde (7 km) and averages over
surrounding grid cells, as a function of the horizontal average. Given the size of a satellite footprint, this
curve provides the spatial averaging error that arises from averaging over footprints.](image)

**Analysis**
The ensemble is analysed using the available observations. Since the observation errors are supposed to be uncorrelated, the observations can be analysed one-by-one. For observation $l$, the first step in the analysis is to compute a gain matrix:

$$ K_l = \tilde{P} f H^T \left( H \tilde{P} f H^T + r_l \right)^{-1} $$

where $r_l$ is the diagonal element of $R$ for observation $l$; the time indices are omitted to simply the notations. Since only one observation is analysed here, the inverse at the right hand side is simply a division by a scalar. The matrix $\tilde{P} f$ is a covariance matrix computed from the ensemble. Simply using the ensemble covariance for this is undesirable, since the spatial correlations represented by the ensemble are usually too strong. This exaggeration is mainly caused by the use of a finite ensemble size, which is useful to represent the major part of the covariance but is unable to represent all details correctly. In addition, the uncertainty model for the emissions has no spatial variation and therewith introduces an artificial increase of spatial correlations. Thirdly, even though their emission origin and timing may be independent, a similar diurnal cycle in the concentrations can create artificial or spurious correlations. As a consequence, the ensemble covariance may suggest that grid cells which are far apart from each other in the domain are strongly correlated, which is undesirable since these correlations are used to distribute the residue between an observation and a simulation over the domain. To be able to ignore the spurious correlations, the localization procedure described in [Houtekamer and Mitchell, 2001] is used. In this procedure, the covariance matrix actually used is formed from a Schur product between the ensemble covariance and a correlation matrix with finite band width. An element $\tilde{p}_{ij}$ of this covariance matrix is computed using:

$$ \tilde{p}_{ij} = p_{ij} C(d_{ij}) $$

where $d_{ij}$ is the horizontal distance between the grid cells holding elements $i$ and $j$, and $C(d)$ is a spatial correlation function. An appropriate formulation for $C(d)$ is determined by examining the spatial correlations present in the simulation (Figure 2). At large distances, the correlation factor vanishes and is explicitly set to zero. As a result, the gain matrix is zero for all elements outside this range, which limits the computational costs since only a small part of the full gain needs to be computed.

With the (localized) gain for observation $l$ available, the ensemble members are analysed by:

$$ \xi^a_{j,l} = \xi^a_{j,l-1} + K_l \left( y_l - H_l \bar{x}^a_{l-1} \right) $$

where $\xi^a_{j,l-1}$ is the ensemble member resulting from the analysis of the previous observation, and similar $\bar{x}^a_{l-1}$ is the mean state computed over all previously analysed members; for the first observation, these are set to the forecast entities. The effect of the analysis is that elements of the state are changed following their relation to simulated observations.
Figure 4 Spatial correlation in formaldehyde simulations in the nature run. Each dot represents a correlation between two time series of concentrations from different grid cells, plotted against the spatial distance between the cells. The averages (bars) are used to fit an appropriate spatial correlation function that can be used for the localization procedure.

Observation screening

In case the assimilation system is unable to represent an observation correctly, assimilation could lead to instability of the system. Such instability can occur if the model lacks certain physical parameterizations or if the model is unable to represent a measurement as a result of a mismatch of the model and measurement spatial or temporal resolution. To avoid this, a screening procedure is applied to reject those measurements which cannot be represented correctly by the assimilation system. The screening procedure is taken from [Jarvinen and Unden, 1997]. If the square of the difference between observation and filter mean is much larger than the expected variance of this difference, the observation is rejected. That is:

\[
(y_i - H_i x_f)^2 > \alpha \left( H_i P_f H_i^T + r_i \right)
\]

Equation 10

where the parameter \( \alpha \) is a threshold factor. The rejected observations are flagged to be able to investigate afterwards where the filter is unable to represent them.

Ensemble size

An important parameter in the filter that has not been discussed yet is the ensemble size. In general, the ensemble size should be large enough to represent the covariance structure imposed by model uncertainty and the model physics. In this application, the covariance structure is rather simple, since all uncertainty is described by the local sources, and observations are available regularly over the domain. Experiments showed that for the described application a limited number of 12-15 modes is sufficient. The current assimilation system is based on conclusions from an extensive sensitivity study carried out within the framework of the SMOGPROG project [Swart et al., 2008], a number of 12 is currently used for the operational forecasts.

4.3.3 Error analysis

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4.4 Emission Estimates related to Aerosols

4.4.1 Input data

The input data for the aerosol emission estimation will be based on AOD retrievals of AATSR and MODIS.

4.4.2 Algorithm description

The emission inversion is based on using the SILAM chemistry transport model driven by 4-dimensional variational inversion scheme.

System for Integrated modeLling of Atmospheric coMposition SILAM (Sofiev et al., 2006, 2008) currently includes both Eulerian and Lagrangian advection-diffusion formulations. The Eulerian core used in the current study is based on the transport scheme of Galperin (1999, 2000), which incorporates the horizontal diffusion term and is combined with the extended resistance analogy of Sofiev (2002) for vertical diffusion.

The removal processes are described via dry and wet deposition. Depending on the particle size, mechanisms of dry deposition vary from the primarily turbulent diffusion driven removal of fine aerosols to the primarily gravitational settling of coarse particles (Slinn and Slinn, 1980). The SILAM wet deposition parameterization (Sofiev et al., 2006, Horn et al., 1987, Smith and Clark, 1989, Jylha, 1991) is based on direct observations performed for moderately hydrophobic aerosols. It distinguishes between sub- and in-cloud scavenging by both rain and snow. The particle size dependence of the impaction scavenging is taken into account by increasing the scavenging rate for super-micron particles in relation to their settling velocity.

The system includes a meteorological pre-processor for diagnosing the basic features of the boundary layer and the free troposphere from the meteorological fields provided by numerical weather prediction (NWP) models (Sofiev et al., 2010). Physical-chemical transformation modules of SILAM include several tropospheric chemistry schemes, basic aerosol dynamics, and radioactive decay processes. The system accepts flexible definition of the particle size spectrum, which can be defined for each specific run depending on the application.

The emission inversion is based on a variant of the 4D-Var data assimilation method, which involves iterative minimization of a quadratic cost function depending on the distance of the observations and their modeled counterparts, and on the distance between the emission estimate and the a priori emission inventory. A detailed description of the emission inversion method has been given in Vira and Sofiev (2012). In the following, we present a summary of the method, and its extension into particulate matter.

Let us denote the parameter (such as initial state or emission rate) of interest as $\xi$, and define the model operator $M$ mapping the parameter, or the control variable, to a unique phase-space trajectory $x = M\xi$ defined over some finite time interval referred as the assimilation window.
The vector of observations $y$ corresponds to the model state $x$ via the observation operator $H$: $y = H(x) + \varepsilon$, where $\varepsilon$ is the observation error, which is assumed to be Gaussian.

The maximum likelihood estimate of the parameter $\xi$ is then the value minimising the cost function

$$J(\xi) = \frac{1}{2}(y - Hx)^T R^{-1}(y - Hx) + \frac{1}{2}(\xi - \xi_b)^T B^{-1}(\xi - \xi_b)$$

(1)

The first term penalises the deviation from the observations $y$ whose accuracy is described by the covariance matrix $R$. The prior knowledge of $\xi$ is included in the background value $\xi_b$ and the background error covariance matrix $B$. The second term therefore penalises the deviation from the prior $\xi_b$. The cost function is minimised using iterative numerical algorithms.

The gradient of equation (1) with respect to $\xi$ is

$$J'(\xi) = M^* H^* R^{-1}(y - Hx) + B^{-1}(\xi - \xi_b),$$

(2)

where $M^*$ and $H^*$ are the tangent linear adjoint model and observation operators, respectively. (Marchuk, 1995).

The forward dispersion model corresponding to the operator $M$ and defining the time evolution of the model state is defined by the scalar transport equation

$$\frac{\partial c_n}{\partial t} + \frac{\partial}{\partial x_i}(u_i c_n) = \frac{\partial}{\partial x_i} \mu \frac{\partial c_n}{\partial x_i} + S_n(x,t) + f_n(x,t),$$

(3)

where $c_n$ is a concentration of the $n$-th species, $f_n(x,t)$ is the emission density, and the chemical sources and sinks are included in $S(c,t)$. If the reaction term is linear, i.e. $S(c,t) = kc_n(x,t)$, then the adjoint equation to (3) reads as (Marchuk, 1995)

$$\frac{\partial c_n^*}{\partial t} - \frac{\partial}{\partial x_i}(u_i c_n^*) = \frac{\partial}{\partial x_i} \mu \frac{\partial c_n^*}{\partial x_i} + kc_n^*(x,t).$$

Here $c^*(x,t)$ is the first-order sensitivity of the functional (1) to a concentration perturbation at time $t$. Its solution corresponds to $M^*$ in (2).

Estimating the complete four dimensional emission distribution is impractical due to under-determination of the problem. Therefore, the approach used in this work and shared by previous studies (Yumimoto et al., 2007, Elbern et al., 2007) is to assume a constant relative deviation of the emission intensity from the background rate throughout the assimilation and forecast windows. The adjusted emission rate is written as $f(x,t) = \alpha(x) f_0(x,t)$, where $\alpha(x)$ is to be estimated. The diurnal emission variations are thus not affected by the assimilation. The correction factor $\alpha(x)$ is assumed to be constant along the vertical axis (height). The sensitivity, and, consequently, the gradient of $J(\xi)$ with respect to $\alpha(x)$ (see equation (2)) is obtained by integrating the solution of the adjoint problem (4) over the assimilation window and the vertical extent of the model domain.

The final emission estimates are given by multiplying the prior emission field with the factors $\alpha$. An assimilation window of 24 hours will be used for the GlobEmission aerosol product.
While the system therefore produces the estimates on daily level, better robustness is expected from the monthly averaged values.

The primary source of observational information for the aerosol emission estimates are satellite retrievals of the aerosol optical depth (AOD). The AOD observation operator is defined by the profile of the mass extinction coefficient, which is computed using the Mie theory for a prescribed particle size distribution. The particle growth due to humidity is considered following Sofiev et al., (2011).

The SILAM model currently includes the emission and formation of primary organic and inorganic, as well as secondary inorganic aerosols. However, the computational cost of the emission inversion requires a simplified representation of the heterogeneous chemistry. The emission inversion is therefore carried out in two steps: first, the contribution of non-sulphuric aerosol compounds (NH4+, NO3−) to the total AOD is computed, and subtracted from the observed values. Then, the optimization problem (1) is solved using a reduced model including only natural and anthropogenic primary aerosols and sulphate formation.

### 4.4.3 Error analysis

- **AATSR**

  The uncertainty estimation for the resulting AOD is obtained by propagating the measurement error of the top-of-atmosphere reflectance by applying inverse problem theory. After the result is determined, the uncertainty is computed using a posteriori covariance. The measurement error is taken to be 5% of the measured reflectance and it is assumed to be uncorrelated. (ESA-CCI project)

- **MODIS**

  A metric analysis of the retrieved AOD values is the tolerance which bounds AOD errors. Hyer et al., 2011 define the target accuracy as:

  \[
  \tau_M = \tau_A \pm (0.05 + 0.2\times\tau_A)\tau_{\text{course}}/5
  \]

  Globally, one standard deviation of “very good” data falls within \((0.05 + 0.2\times\tau_A)\) error thresholds (\(\tau_A\) is the AOD at the MODIS retrieval at 555 nm). However, for most applications a prognostic RMS (root mean square) error model with a noise floor is more appropriate. For global applications using MODIS level 2 data over land, we recommend the use of the greater of 0.08 or 0.02 +0.22×\(\tau_M\) for terra and 0.07 or 0.01+0.26×\(\tau_M\) for Aqua. (Shi et al., 2011, Zhang et al 2006, Remer et al 2005, among others). Where \(\tau_M\) is “Corrected Optical Depth-Land” from the level 2 product.

The emission estimates are affected by the uncertainties in observations, a priori emissions as well as uncertainties in the chemistry-transport model. The observational and a priori emission errors are encoded in the covariance matrices \(R\) and \(B\) in Eq. (1). While an algebraic relation between \(R, B,\) and the a posteriori errors is easy to derive, its evaluation is computationally difficult due to the dimension of the problem. Moreover, the effect of forward model errors is not quantified this way.
The sensitivity to errors in the input data will be studied using Monte-Carlo simulations with synthetic observations. The effect of modeling errors can be investigated with cross-validation using several chemistry-transport models with independent inversion schemes.
4.5 Emission Estimates related to fires

4.5.1 Input data

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The emission inversion is based on a variant of the 4D-Var data assimilation method, which involves iterative minimization of a quadratic cost function depending on the distance of the observations and their modeled counterparts, and on the distance between the emission estimate and the a priori emission inventory. A detailed description of the emission inversion method has been given in Vira and Sofiev (2012). In the following, we present a summary of the method, and its extension into particulate matter.

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The vector of observations $y$ corresponds to the model state $x$ via the observation operator $H$: $y = H(x) + \varepsilon$, where $\varepsilon$ is the observation error, which is assumed to be Gaussian.

The maximum likelihood estimate of the parameter $\xi$ is then the value minimising the cost function

$$J(\xi) = \frac{1}{2} (y - Hx)^T R^{-1} (y - Hx) + \frac{1}{2} (\xi - \xi_b)^T B^{-1} (\xi - \xi_b)$$

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The first term penalises the deviation from the observations $y$ whose accuracy is described by the covariance matrix $R$. The prior knowledge of $\xi$ is included in the background value $\xi_b$ and the background error covariance matrix $B$. The second term therefore penalises the deviation from the prior $\xi_b$. The cost function is minimised using iterative numerical algorithms.

The gradient of equation (1) with respect to $\xi$ is

$$J'(\xi) = M^* H^* R^{-1} (y - Hx) + B^{-1} (\xi - \xi_b),$$

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where $M^*$ and $H^*$ are the tangent linear adjoint model and observation operators, respectively. (Marchuk, 1995).

The forward dispersion model corresponding to the operator $M$ and defining the time evolution of the model state is defined by the scalar transport equation

$$\frac{\partial c_n}{\partial t} + \frac{\partial}{\partial x_j} (u_j c_n) = \frac{\partial}{\partial x_i} \mu_{ij} \frac{\partial c_n}{\partial x_l} + S_n(x,t) + f_n(x,t),$$

(3)

where $c_n$ is a concentration of the $n$-th species, $f_n(x,t)$ is the emission density, and the chemical sources and sinks are included in $S(c,t)$. If the reaction term is linear, i.e. $S(c,t) = kc_n(x,t)$, then the adjoint equation to (3) reads as (Marchuk, 1995)

$$\frac{\partial c_n^*}{\partial t} - \frac{\partial}{\partial x_i} (u_j c_n^*) = \frac{\partial}{\partial x_j} \mu_{ij} \frac{\partial c_n^*}{\partial x_j} + kc_n^*(x,t).$$

Here $c_n^*(x,t)$ is the first-order sensitivity of the functional (1) to a concentration perturbation at time $t$. Its solution corresponds to $M^*$ in (2).

Estimating the complete four dimensional emission distribution is impractical due to under-determination of the problem. Therefore, the approach used in this work and shared by previous studies (Yumimoto et al., 2007, Elbern et al., 2007) is to assume a constant relative deviation of the emission intensity from the background rate throughout the assimilation and forecast windows. The adjusted emission rate is written as $f(x,t) = \alpha(x) f_0(x,t)$, where $\alpha(x)$ is to be estimated. The diurnal emission variations are thus not affected by the assimilation. The correction factor $\alpha(x)$ is assumed to be constant along the vertical axis (height). The sensitivity, and, consequently, the gradient of $J(\xi)$ with respect to $\alpha(x)$ (see equation (2)) is obtained by integrating the solution of the adjoint problem (4) over the assimilation window and the vertical extent of the model domain.

The final emission estimates are given by multiplying the prior emission field with the factors $\alpha$. An assimilation window of 24 hours will be used for the GlobEmission aerosol product.
While the system therefore produces the estimates on daily level, better robustness is expected from the monthly averaged values.

The primary source of observational information for the aerosol emission estimates are satellite retrievals of the aerosol optical depth (AOD). The AOD observation operator is defined by the profile of the mass extinction coefficient, which is computed using the Mie theory for a prescribed particle size distribution. The particle growth due to humidity is considered following Sofiev et al., (2011).

The SILAM model currently includes the emission and formation of primary organic and inorganic, as well as secondary inorganic aerosols. However, the computational cost of the emission inversion requires a simplified representation of the heterogeneous chemistry. The emission inversion is therefore carried out in two steps: first, the contribution of non-sulphuric aerosol compounds (NH4+, NO3-) to the total AOD is computed, and subtracted from the observed values. Then, the optimization problem (1) is solved using a reduced model including only natural and anthropogenic primary aerosols and sulphate formation.

### 4.5.3 Error analysis

- **AATSR**

The uncertainty estimation for the resulting AOD is obtained by propagating the measurement error of the top-of-atmosphere reflectance by applying inverse problem theory. After the result is determined, the uncertainty is computed using a posteriori covariance. The measurement error is taken to be 5% of the measured reflectance and it is assumed to be uncorrelated. (ESA-CCI project)

- **MODIS**

A metric analysis of the retrieved AOD values is the tolerance which bounds AOD errors. Hyer et al., 2011 define the target accuracy as:

\[
\tau_M = \tau_A \pm (0.05 + 0.2 \times \tau_A) \tau_{course}
\]

Globally, one standard deviation of “very good” data falls within \((0.05 + 0.2 \times \tau_A)\) error thresholds \((\tau_A\) is the AOD at the MODIS retrieval at 555 nm). However, for most applications a prognostic RMS (root mean square) error model with a noise floor is more appropriate. For global applications using MODIS level 2 data over land, we recommend the use of the greater of 0.08 or 0.02 +0.22×\(\tau_M\) for terra and 0.07 or 0.01+0.26×\(\tau_M\) for Aqua. (Shi et al., 2011, Zhang et al 2006, Remer et al 2005, among others). Where \(\tau_M\) is “Corrected Optical Depth-Land” from the level 2 product.

The emission estimates are affected by the uncertainties in observations, a priori emissions as well as uncertainties in the chemistry-transport model. The observational and a priori emission errors are encoded in the covariance matrices \(R\) and \(B\) in Eq. (1). While an algebraic relation between \(R, B\), and the a posteriori errors is easy to derive, its evaluation is computationally difficult due to the dimension of the problem. Moreover, the effect of forward model errors is not quantified this way.
The sensitivity to errors in the input data will be studied using Monte-Carlo simulations with synthetic observations. The effect of modeling errors can be investigated with cross-validation using several chemistry-transport models with independent inversion schemes.
4.6 Emission Estimates related to fires

4.6.1 Input data

The Integrated System for wild-land FIRES (IS4FIRES) v.1.2 is based on Level 2 MODIS Collection 5.1 Active Fire Products, which are used for the near-real-time and historical evaluation of the emissions from wild-land fires, as well as a series of other input data (Figure 5). Considerations are given to include fire products of VIIRS, GOES-2 and GOES-3 when these data become available.

![Figure 5. Scheme of data flows of IS4FIRES v.1.2.](image)

The incorporation of the fire injection profile resulted in a strict separation of the satellite-related data containing the fire information and land cover from the meteorology-related data used for calculation of the initial 3D plume distribution. For the sake of efficiency, the second part has been incorporated inside the SILAM model – because this is the first system that naturally involves the meteorological–data processing routines. Therefore, the IS4FIRES v.1.2 per-se became smaller: it only checks, reprojects and aggregates the MODIS fire observations in the daily fire observations. The rest of emission calculation takes place in SILAM, which calculates the absolute emission and its 3D injection clouds dynamically while evaluating the impact of fires on atmospheric composition and air quality.
The present IS4Fires system consists of two parallel branches based on partly independent products: the Temperature Anomaly and Fire Radiative Power. Their algorithms of converting the fire information to the emission fluxes of atmospheric pollutants are described below, starting from the outlines of the corresponding fire products. They are used so that, if the FRP information on the specific fire is available, it is taken as-is. Should the FRP processing of MODIS fail but the TA processing pass, the TA value is used for producing the would-be-FRP value following the non-linear regression suggested by Sofiev et al, (2009).

The MODIS fire detection procedure is based on a contextual algorithm of Giglio et al (2003) that exploits the strong emission of mid-infrared radiation from fires (Dozier, 1981; Matson and Dozier, 1981). The algorithm examines each pixel of the MODIS swath and attributes it to one of the following classes: missing data, cloud, water, non-fire, fire, or unknown. For each fire-classified pixel, the procedure attempts to use the neighbouring pixels to estimate the radiometric signal of the pixel, if there would be no fire there. Valid neighbouring pixels are identified in a window centred on the potential fire pixel and used to estimate this background value.

4.6.2 Algorithm description

The procedure of determining the emission from the wild-land fires will generally follow that of aerosol products – with a few exceptions. Firstly, the background emission will be taken from IS4Fires system of FMI outlined below. Secondly, the data assimilation will be used to constrain both particulate matter and gaseous species, with primary attention paid to CO.

The Fire Radiative Power (FRP, a rate of release of Radiation Energy, FRE) of the fire pixel is based on the empirical formula of Kaufman et al (1998):

\[
FRP = 4.34 \times 10^{-13} (T_4^8 - T_{4b}^8) \text{ [Watt]},
\]

where the \(T_4\) and \(T_{4b}\) are the fire and the background (taken from neighbouring pixels) temperatures, respectively, measured at the 4-\(\mu\)m channel. The dependence has been obtained from fitting the actual release of radiative energy from a fire and its apparent temperature at the 4 and 11 \(\mu\)m channels – as observed by the MODIS instrument. The relationship showed good correlation for open moderate-to-strong fires (Kaufman et al, 1998). There may be potential difficulties for small fires, as these may be partly overshadowed by trees, appear as low-temperature but strongly emitting smouldering fires, etc.

Both TA and FRP data are included into the level 2 Fire Products (MOD14 for Terra and MYD14 for Aqua satellites).

To convert the FRP to emission fluxes we used a similar approach as for TA – a direct conversion of FRP using an empirical scaling to emission rates. In the current FAS it is based on Ichoku & Kaufman (2005, hereinafter referred as IK05) who related the FRP in [W] per pixel to total particulate matter (PM) emission in [kg tPM s\(^{-1}\)]. Since the calibration IK05 was obtained by relating the aerosol optical depth (AOD) with the FRP, the obtained emission factors are valid for total PM instead of PM\(_{2.5}\), which was the reference species for FAS-TA. The mean relation between these PM measures can be evaluated based on AM01: within the fire plume.
The relation is approximately valid for all land use types: the changes between the vegetation types are smaller than the uncertainty range within each type (AM01).

The key parameter for IS4FIRES is therefore the emission rate of total PM per unit FRP, i.e. the smoke emission factor $C_e$ [kg tPM J⁻¹]. According to IK05, $C_e$ varies from 0.02-0.06 kg tPM MJ⁻¹ for boreal regions, 0.04-0.08 kg tPM MJ⁻¹ for Africa (mainly savannas and grassland), and 0.08-0.1 kg tPM MJ⁻¹ for Western Russian regions. Since the $C_e$ determination involved a simple estimate of atmospheric transport (based on wind at a constant height and not involving a dispersion model), the authors suggested that the coefficients are probably overestimated by about a factor of 2. Using these estimates as a starting point, we have re-estimated the emission coefficients using actual land-cover information, instead of geographical regions. For the IS4FIRES v.0.99 and v.1.0, three land cover classes were considered: forest, grass, and mixture of both. The corresponding total-PM emission factors were suggested as: 0.035 kg tPM MJ⁻¹ for forest, 0.018 kg tPM MJ⁻¹ for grassland and agriculture, and 0.026 kg tPM MJ⁻¹ for mixed areas. These values were deduced from the prevailing land cover in the IK05 domains.

For the IS4FIRES v.2, the primary deliverable of the GlobEmission, the list of land cover classes was increased to 7 types (Table 2).

<table>
<thead>
<tr>
<th>Land cover</th>
<th>PM₂.₅ factor</th>
<th>unit</th>
<th>PM₁₀ factor</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tropical forest</td>
<td>0.00697</td>
<td>kg/MJ</td>
<td>0.01417</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Grass</td>
<td>0.00410</td>
<td>kg/MJ</td>
<td>0.00474</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Crop residue</td>
<td>0.01003</td>
<td>kg/MJ</td>
<td>0.02083</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Pasture maintenance</td>
<td>0.02058</td>
<td>kg/MJ</td>
<td>0.04020</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Boreal forest</td>
<td>0.01085</td>
<td>kg/MJ</td>
<td>0.01419</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Temperate forest</td>
<td>0.00410</td>
<td>kg/MJ</td>
<td>0.00484</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Peat</td>
<td>0.02086</td>
<td>kg/MJ</td>
<td>0.04173</td>
<td>kg/MJ</td>
</tr>
<tr>
<td>Shrub</td>
<td>0.00475</td>
<td>kg/MJ</td>
<td>0.00558</td>
<td>kg/MJ</td>
</tr>
</tbody>
</table>

We assume that inside the fire plumes, the AOD was entirely determined by the biomass-burning products. Thus, Saarikoski et al (2007) found that more than 80% of PM₂.₅ during a specific episode in May 2006 was originated from fires – in comparatively aged plumes. We
therefore attributed all systematic discrepancy between the observed and calculated column AOD to errors in the emission rates, and corrected the emission factors accordingly.

The intermediate calibration performed on the basis of a single year 2008 resulted in the emission factors shown in Table 2. For the final calibration, these values will be used as a starting point for refinement via both scaling calibration for other years and variational assimilation over specific episodes and regions, especially those dominated by a specific land cover type.

### 4.6.3 Error analysis

Direct error analysis of emission estimates is not possible due to no direct observations of this quantity. Therefore, the error analysis will concentrate on two complementary directions: (i) evaluation of the resulting model plumes from fires using the satellite and in-situ pollutant data, (ii) inter-comparison with existing emission inventories for the same time period.

Comparison with existing inventories will be made for evaluating the general uncertainties of the emission estimates rather for correcting the GlobEmission estimates: a-priori it is not known, which inventory is more accurate.

Provision will be given to application of the obtained emission inventory in another model of GlobEmission – LOTOS-EUROS, which would demonstrate the impact of the model formulations to the emission estimations.
4.7 High Resolution Emission Inventories

4.6.1 Input data

- **Regional emission inventory for Middle East, provided by KNMI**: KNMI will provide emission estimates of NOx on a 0.25 degree resolution, derived from satellite observations and inverse modelling, for the Middle East.

- **Sector split provided by QEERI or based on literature search**: The regional emission inventory for the Middle East will be provided by KNMI as total emissions, i.e. without any breakdown over different economic sectors. As further downscaling is highly dependent on the breakdown over different sectors, information on the sectoral split is required.

  For South Africa (GlobEmission, phase 1), relative values were taken from literature, however these data had a lot of drawbacks: the data were outdated, they were only available at national level, only dealt with anthropogenic emissions, and only 4 main sectors were included, … .

  For the downscaling application in Qatar, we hope to find better data. Hereo, it was agreed with QEERI, the GlobEmission user from Qatar, that VITO will perform a literature search on available data with respect to relative contributions (biogenic versus anthropogenic on one hand and contributions of (sub)sectors within both, biogenic and anthropogenic emissions, on the other hand). Moreover, VITO will perform a screening on the availability of similar data in surrounding areas (Saudi Arabia, UAE, … ). QEERI from its side will investigate which data are available via the Qatar national reporting system (Qatar needs to report emissions to the UN). Furthermore, it was also agreed that QEERI will facilitate discussions with eventual data suppliers (e.g. from surrounding areas) on VITOs request.

  As a back-up plan (in case no useful data will be found through the above mentioned channels), EDGAR emission estimates will be used, i.e. the relative contributions from the EDGAR estimates will be applied on the NOx estimates provided by KNMI. This however requires that the sector list that will be taken into account with respect to the downscaling for Qatar, somehow can be matched to the EDGAR sector classification.

- **Proxy data provided by QEERI**: According to QEERI, the most important sources with respect to NOx emissions in Qatar, can be found within the following sectors:

  → Anthropogenic sources:
    - Oil and Gas
    - Off shore Oil and Gas
    - Water desalination & Power generation
    - Road Transport
    - Shipping
    - Aviation
Residential & Commercial
Agriculture, Farms?

→ Biogenic emissions

Consequently, good proxy data for each of these sectors are required.

Agreements on data search and delivery were already made with QEERI. As ultimate deadline, the end of May 2015 was proposed. This would allow VITO to develop and implement a downscaling prototype system, which can be presented on the User Workshop, foreseen in October 2015. Based on the feedback received during the workshop, the prototype system will be adapted during the last project period (October 2015 – March 2016).

4.6.2 Algorithm description

The methodology to downscale NOx emissions for Qatar strongly depends on the input data that will be received. Therefore, it is difficult to describe an Algorithm Theoretical Baseline (ATB) at this stage. However, we are quite confident that useful data will be provided and therefore strongly assume that the methodology developed for South Africa can be taken as a starting point. Below, the methodology used for downscaling of South African NOx emissions is outlined (note that it strongly correlates to the data that were available for South Africa).

The methodology for downscaling the regional inventory consisted of the following steps:

1. The gridded regional (0.25 degree resolution) emission estimates from KNMI were added up to a national emission total for South Africa, based on satellite observations;

2. Spatial patterns were generated both, on a 0.25x0.25 degrees grid (step 2a) and on a 0.05x0.05 degrees grid (step 2b) using the available geographical proxy data, i.e. roads, population density, land use data, … in order to
   a. “reconstruct” the low resolution 0.25x0.25° KNMI emissions map (see step 3),
   b. allow further downscaling to high resolution (0.05x0.05°, see step 4);

3. Reconstruction of the low resolution emissions map from
   a. the national emissions total for South Africa (step 1) and
   b. the spatial patterns on the original 0.25x0.25 degrees grid (step 2a),

making use of an ‘optimization procedure’. An optimized linear combination of the different spatial patterns was determined. Hereto, validation indicators between the original map derived from inverse modelling and the reconstructed map were used as cost function for the optimization (minimization):
where \( r_{xy(a)} \) is the spatial correlation coefficient between the original grid values \( x_i \) (as derived by the inverse modelling) and the reconstructed grid values \( y_i(a) \) (as calculated using \( a \) as a weight vector between de different proxy parameters).

Via maximization of the spatial correlation, the methodology tries to explain as much as possible of the spatial variance of the original KNMI emission dataset through a linear combination of the proxy-patterns.

Key to the methodology is therefore:

i. determination of relative importance of each sector
ii. at national level
iii. through optimization of weights

4. **Validation and optimization of the minimization procedure.** Different minimization techniques, i.e. based on three constraining modes, were applied and compared against each other. The three constraining modes that were taken into account can be summarized as follows:

a. **Free:** where the only constraint that applies is that the sum of the weights should equal 1 and the weights should all be non-negative.

b. **Hard constraint:** where, next to being non-negative, 4 additional constraints guaranteeing that the explicit sector split known for anthropogenic emissions is maintained, are built in. These constraints are:
   i. The traffic proxy weight should equal 30.17 % of \((1 – \text{the sum of the biogenic emission weights})\)
   ii. The residential proxy weight should equal 6.16% of \((1 – \text{the sum of the biogenic emission weights})\)
   iii. The industry proxy weight should equal 8.92 % of \((1 – \text{the sum of the biogenic emission weights})\)
   iv. The powerplant weight should equal 5.47 + 1.28 % of \((1 – \text{the sum of the biogenic emission weights})\). The remaining 1.28 % of the above pie-chart was added in order to make the different anthropogenic proxies add up to a total of 100 % for anthropogenic emissions.

c. **Soft constraint:** where the same methodology as in the hard constraints mode is adapted, however, allowing the different contributions of the anthropogenic emissions to vary within +/- 5 % of the split given above.

5. **Computation of high resolution emission maps from**

a. the national emissions total for South Africa (step 1)

b. the spatial patterns on the high resolution 0.05x0.05 degrees grid (step 2b)

c. the optimized weights, i.e. vector \( a \) in the above formula (step 3)
6. Note to the algorithm: national totals (as derived from original KNMI data) are conserved in the algorithm. However, locally there might be differences due to the nature of the approach. Hence, a local residue correction scheme might be applied as a last step.

The fact that the core of the methodology was developed during GlobEmission, phase 1, and that the existing methodology most probably can be taken as a starting point to develop a downscaling methodology for Qatar, leaves room to focus on fine tuning of the methodology. For instance, attempts will be made to improve on the balance between anthropogenic and biogenic emissions and to better conserve the local mass balance in the downscaling. Furthermore we will adapt the methodology to the Qatar specific situation (e.g. dealing with off-shore and shipping emissions in the Gulf).

4.7.1 Error analysis

The optimization procedure is part of the proposed downscaling methodology. Hence, an important part of the error analysis makes inherently part of the proposed methodology. The application of adaptations to the main input parameters in order to minimize the scaling factors is intended to reduce the uncertainty of the downscaled emissions.