MEGAPOLI Scientific Report 11-08

Effective Emission Factors for OC and BC for Urban Type Emissions

MEGAPOLI Deliverable D3.3

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The French SAFIRE ATR-42 research aircraft (Météo-France-CNRS-CNES) at the Cergy Pontoise airport during the MEGAPOLI summer campaign.

2011
Colophon

Serial title:
MEGAPOLI Scientific Report 11-08

Title:
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Subtitle:
MEGAPOLI Deliverable D3.3

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Language:
English

Keywords:
Source apportionment, chemical composition, urban, suburban, variability

Url:
http://megapoli.dmi.dk/publ/MEGAPOLI_sr11-08.pdf

Digital ISBN:
978-87-92731-12-8

MEGAPOLI:
MEGAPOLI-34-REP-2011-06

Website:
www.megapoli.info

Copyright:
FP7 EC MEGAPOLI Project
Content:

Abstract ................................................................................................................................................4
1. Introduction......................................................................................................................................5
2. Methodology ....................................................................................................................................5
3. Input Data and Model Description...................................................................................................6
   3.1. MEGAPOLI measurements ......................................................................................................6
   3.2. TNO emissions inventories ......................................................................................................6
   3.3. PM speciation factors ................................................................................................................8
   3.4. CHIMERE chemistry-transport model .....................................................................................9
   3.5. Input data, model configuration and domains..........................................................................9
   3.6. Simulated cases .......................................................................................................................10
   3.7. Procedure to Compute Emission Factors ....................................................................................11
4. Results and Discussions.................................................................................................................13
   4.1. Emission evaluation factors ....................................................................................................13
   4.2. Benefits and uncertainties of the methodology ........................................................................15
   4.3. Mixing layer depth adjustment ..............................................................................................15
   4.4. Case of the 21st of July ...........................................................................................................17
   4.5. Hourly emission contribution to plume ..................................................................................20
   4.6. Deposition ...............................................................................................................................21
   4.7 Other errors ..............................................................................................................................22
Conclusions........................................................................................................................................23
Acknowledgements............................................................................................................................24
References..........................................................................................................................................25
Previous MEGAPOLI reports............................................................................................................26
Abstract

The estimation of pollutant emissions in megacities is of major concern for the design of effective air pollution abatement strategies. Within the framework of FP7 EC MEGAPOLI project, an intensive campaign of both airborne and ground-based pollutant measurements was carried out in the Paris region in July 2009 and January-February 2010 to quantify the sources of organic aerosol and to improve our understanding of the secondary aerosol formation.

The aim of this report is to evaluate two emissions inventories delivered by TNO, without and with refined treatment of Paris emissions from AIRPARIF data, designated as MEGAPOLI and MEGAPOLI-MC inventories, respectively. A third inventory designated as EMEP-LA, consisting of BC and OC emissions from the Laboratoire d’Aérologie inventory and other anthropogenic emissions from the EMEP inventory is also considered.

An original method based on the use of airborne measurements is developed to evaluate these emission inventories. Several chemically inert tracer species are considered, NOy and BC, for which plume measurements are available from the French ATR-42 aircraft, to evaluate NOx and BC emissions respectively. High resolution 3x3 km simulations with the regional chemistry transport model CHIMERE, coupled with the above inventories are used to simulate tracer concentrations in the plume. For all flights and simulations, the Paris plume can be well discriminated from the background. For both observations and simulations, along the flight path perpendicular to the plume, the plume integral of tracer concentration above background is calculated, giving for each flight spatially averaged emission correction factors for the Paris region. The method allows reducing some potential errors due to model uncertainties (plume dilution, horizontal advection, and vertical dispersion). POA emission factors have not been considered as initially planned, because reliable POA measurements are not yet available for the summer campaign (mainly due to the rather low POA values encountered during the summer campaign).

The results suggest a potential overestimation of NOx emissions for all three inventories. BC emissions may be also overestimated by the EMEP-LA and MEGAPOLI inventories, but may be underestimated by MEGAPOLI-MC. However, results show a high day-to-day variability, leading to significant uncertainties. Several uncertainty sources have been investigated in order to evaluate on the one hand the emission variability, and on the other hand the uncertainties due to the method itself. Thus, emission correction factors are in general different from unity at approximately a one sigma confidence level.

The main uncertainty sources are: (i) the wind speed that directly links the diurnal emission profile to the rate of decrease of concentrations in the plume, (ii) the degree of vertical mixing that determines the representativeness of the airborne measured concentrations, (iii) the wet and dry deposition of the tracers which can lead to discrepancies in the emission factors if these processes are not well simulated in the model, and (iv) the boundary layer height and its horizontal variability over the aircraft trajectory which directly affects the level of concentrations.

The availability of evaluated and corrected emissions inventories is of great importance for assessing pollution transformation processes within the MEGAPOLI project. The developed methodology is of general interest for evaluating megacity emissions.
1. Introduction

Accurate pollutant emission inventories in megacities are needed for the improvement of air pollution modelling. Within the framework of MEGAPOLI, highly resolved emission inventories have been built by TNO over Europe without and with refined treatment of megacity emissions. In parallel, an intensive campaign of both airborne and ground-based pollutant measurements was carried out in the Paris region in July 2009 and January-February 2010 with the major objective to quantify the sources of organic aerosol and to improve our understanding of secondary aerosol formation.

The aim of this study is to evaluate emission inventories in the Paris region. An original method based on the use of airborne measurements is developed to perform this task. The method reduces errors due to model uncertainties (plume dilution, horizontal advection, vertical dispersion), and is applicable to quasi inert tracers and emission totals averaged over the agglomeration. In our case, BC and NOy will be considered to evaluate BC and NOx emission inventories respectively; the method should also work with quasi inert VOCs. At this point, our study is focused on the summer MEGAPOLI campaign during which eleven air-borne measurement flights have been performed. POA emission factors have not been considered as initially planned, because reliable POA measurements are not yet available for the summer campaign (mainly due to the rather low POA values encountered during the summer campaign).

The paper is structured as follows. Section 1 explains the methodology. In Section 2, all required input is described, including the required measurements during the MEGAPOLI summer campaign, the emission inventories delivered by TNO, the factors speciating the particulate matter emissions into BC and OC, the CHIMERE chemistry-transport model used for simulations with these inventories, and characteristics of the simulations performed. Section 3 explains in detail the procedure applied to compute the emission factors. Section 4 shows and discusses the results, considering the various sources of uncertainty.

2. Methodology

The method developed in this study aims to evaluate several chemically inert tracer species (such as NOy and black carbon) emission inventories over a megacity. It is based on airborne measurements of these species in the megacity plume during the afternoon in a well-mixed convective boundary layer, so that the vertical mixing can be considered as established, and consequently the measured concentrations at a particular altitude as representative of the concentrations in the whole mixing layer.

A simulation with the CHIMERE chemistry-transport model (CTM), coupled with the inventories to be evaluated, is used to simulate tracer concentrations in the plume. For both observations and simulations, along the flight path perpendicular to the plume, the plume integral of tracer concentration above background can be calculated, yielding, through the ratio of simulated area over measured area, spatially averaged emission correction factors for the Paris region for each flight.

By considering integrated peak areas over lateral transects through the plume, such a method allows to avoid some potential errors in the structure of the simulated plume, e.g., errors in wind direction, any error on lateral dispersion, etc., and consequently to focus on emissions. However, several potential error sources still remain, and therefore need to be carefully considered: (i) the wind speed which directly links the diurnal emission profile to the rate of decrease of concentrations in the plume, (ii) the degree of vertical mixing which determines the representativeness of the airborne measured concentrations, (iii) the wet and dry deposition of the tracers which can lead to discrepan-
cies in the emissions factors if not well simulated in the model, and (iv) the boundary layer height and its horizontal variability over the aircraft trajectory which directly affects the level of concentrations.

3. Input Data and Model Description

3.1. MEGAPOLI measurements

Both meteorological and chemical observations, obtained during the MEGAPOLI summer campaign in July 2009 have been used in this study. Among the chemical data available, both NOy and BC airborne measurements from the French ATR-42 aircraft have been used.

NOy concentrations were measured at a 30 s time resolution with an analyser from Ecophysics in which NO is measured using ozone chemiluminescence. NO₂ is photolytically converted, and NOy is converted with gold in a heated oven (Credit: Aurélie Colomb, LaMP, Vincent Michoud, LISA). The limit of detection is 10 pptv. NO, NO₂ and NOy measurement uncertainties are 10, 20 and 20%, respectively. According to Ecophysics, the measured NOy includes the following species: NO, NO₂, HNO₂, HNO₃, NO₂NO₂, N₂O₅, PAN, PPN and particulate nitrate. BC concentrations are provided with a 1 s time resolution from the light absorption coefficient at 650 nm with the PSAP instrument, using a mass specific absorption coefficient QBC of 6.6 m²/g (Credit: Alfons Schwarzenboeck, Suzanne Crumeyrolle, LaMP). Airborne NOy and BC measures are available for several days in July: 1st, 9th, 10th, 13th, 15th (only BC that day), 16th, 20th, 21st, 25th, 28th, 29th (only NOy that day).

Various physical parameters were also measured in the ATR-42 aircraft at a 1 s time resolution among which wind speed, wind direction and position of the aircraft (longitude, latitude, height) have been also used. Wind measurements were provided by several instruments (dynamic pressure, GPS) on the SAFIRE platform. Among other meteorological data available, mixing layer depths at SIRTA (suburban background site) and LHVP (urban background site) have also been used. At SIRTA, continuous mixing layer depths are estimated from ALS450 Leosphere backscatter lidar data at a 5min time resolution (Credit: Yoshann Morille, LMD). At LHVP, values are estimated from CL31 ceilometer data. Other meteorological parameters have been also used to evaluate the quality of the meteorological input data in our chemistry model. More precisely, we used Leosphere lidar wind cube measurements at SIRTA which provided wind measurements at a 10 min time resolution, each 20 m from 40 to 200 m of height (Credit: Jean-Charles Dupont, LMD).

3.2. TNO emissions inventories

In the framework of the FP7 MEGAPOLI project, TNO has provided two highly resolved emission inventories over Europe for SO₂, NOx, NH₃, CO, NMVOC, CH₄, PM2.5 and PM10, including emissions in more than 40 countries.

Using a top-down approach, the first inventory described in MEGAPOLI Deliverable D1.2 was built using various data sources: official emissions submitted by European countries¹, emissions calculated with the IIASA’s RAINS model² in order to fill eventual gaps, own TNO data, data from the CEPMEIP³ database and expert judgments. The second inventory described in MEGAPOLI Deliverable D1.6 includes refined local emission data over two European megacities (Paris, London)
and two urban conglomerations (Po Valley in Italy, Rhine-Ruhr region in Germany, later designated as megacities, too) using a bottom-up approach: for these megacities or regions, detailed local inventories compiled by local agencies are available: AIRPARIF for Paris, GLA (Greater London Authority) for London, the NRW (North Rhine-Westphalia) regional inventory for Rhine-Ruhr, and municipal level inventories of four Italian regions (Piemonte, Lombardia, Friulu Venezia Giulia and Trentino Alto Adige) based on INEMAR methodology for the Po Valley.

Emissions are detailed by SNAP sectors, according to SNAP 97 1st level nomenclature (Table 1). Emissions from area sources have been spatially distributed according to eleven source sector specific distribution proxies.

### Table 1: SNAP sectors description.

<table>
<thead>
<tr>
<th>SNAP sector</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Public electricity and other energy transformation</td>
</tr>
<tr>
<td>2</td>
<td>Small combustion plants</td>
</tr>
<tr>
<td>3</td>
<td>Industrial combustion and processes with contact</td>
</tr>
<tr>
<td>4</td>
<td>Industrial process emission</td>
</tr>
<tr>
<td>5</td>
<td>Fossil fuel production</td>
</tr>
<tr>
<td>6</td>
<td>Solvent and product use</td>
</tr>
<tr>
<td>7</td>
<td>Road transport</td>
</tr>
<tr>
<td>7.1</td>
<td>Road transport Gasoline</td>
</tr>
<tr>
<td>7.2</td>
<td>Road transport Diesel</td>
</tr>
<tr>
<td>7.3</td>
<td>Road transport LPG</td>
</tr>
<tr>
<td>7.4</td>
<td>Non-exhaust (volatilization)</td>
</tr>
<tr>
<td>7.5</td>
<td>Non-exhaust (brake wear, tire wear, road wear)</td>
</tr>
<tr>
<td>8</td>
<td>Other (non-road) transport and mobile machinery</td>
</tr>
<tr>
<td>9</td>
<td>Waste disposal</td>
</tr>
<tr>
<td>10</td>
<td>Agriculture</td>
</tr>
<tr>
<td>11</td>
<td>Nature*</td>
</tr>
</tbody>
</table>

* Not prepared in these emissions inventories

MEGAPOLI Deliverable D1.6 has analyzed significant differences between the local bottom-up and down-scaled regional emission inventories. The ratios for the different species are recalled in Table 2. In general, ratios are larger than unity. For Paris, both inventories are quite equivalent for NOx emissions, but the regional one gives stronger emissions for all the other species, particularly for the PM$_{10}$ and PM$_{2.5}$ emissions which are about three times larger. In the second inventory provided by TNO, local emissions are retained over megacities, but national emissions (outside of the megacities are adjusted (increased) to let consistent national totals.

### Table 2: Ratio of megacities emissions to regional European scale inventory over local megacities emission inventory /source: MEGAPOLI deliverable D1.6, page 8/.

<table>
<thead>
<tr>
<th>Megacity</th>
<th>NOx</th>
<th>PM$_{10}$</th>
<th>PM$_{2.5}$</th>
<th>NH$_3$</th>
<th>SO$_2$</th>
<th>NMVOC</th>
<th>CO</th>
</tr>
</thead>
<tbody>
<tr>
<td>London</td>
<td>1.6</td>
<td>3.9</td>
<td>n.a.</td>
<td>n.a.</td>
<td>9.4</td>
<td>1.5</td>
<td>3.6</td>
</tr>
<tr>
<td>Paris</td>
<td>1.1</td>
<td>3.0</td>
<td>3.2</td>
<td>2.2</td>
<td>1.9</td>
<td>1.9</td>
<td>4.1</td>
</tr>
<tr>
<td>Rhine-Rhur</td>
<td>0.9</td>
<td>1.6</td>
<td>1.2</td>
<td>1.6</td>
<td>1.1</td>
<td>2.4</td>
<td>0.7</td>
</tr>
<tr>
<td>Po Valley</td>
<td>0.9</td>
<td>0.9</td>
<td>n.a.</td>
<td>1.0</td>
<td>1.2</td>
<td>0.8</td>
<td>1.4</td>
</tr>
</tbody>
</table>

* Urban/rural population, land use, coastal waterways, high pressure gas network, inland waterways, non-urban road network, European Rail network, distribution of cattle, distribution of pigs, distribution of poultry, distribution of other animals.
The final spatial resolution of both inventories is 1/8° in longitude and 1/16° in latitude (roughly 7 x 7 km), to be compared to the 50x50 km spatial resolution of EMEP inventory which was a reference for European air quality modelling before. Furthermore, the MEGAPOLI-MC inventory also provides higher resolutions over megacity domains: 1 x 1 km for London and Paris region, 1/60° x 1/60° (roughly 1 x 2 km) for the Rhine-Ruhr area, 4 x 4 km for Po Valley.

### 3.3. PM speciation factors

For PM$_{2.5}$ and PM$_{10}$ emissions included in both inventories, speciation factors are provided by TNO for each country and each SNAP sector to get POA (primary organic aerosol) and BC (black carbon) parts, both for the PM$_{2.5}$ and PM$_{10}$ size classes. Speciation factors for France are given in the Tables 3 and 4.

**Table 3: PM$_{10}$ speciation factors for each SNAP sector.**

<table>
<thead>
<tr>
<th>SNAP sector</th>
<th>BC [%]</th>
<th>POA [%]</th>
<th>Other* [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10.3</td>
<td>2.3</td>
<td>87.4</td>
</tr>
<tr>
<td>2</td>
<td>17</td>
<td>72.5</td>
<td>10.5</td>
</tr>
<tr>
<td>3</td>
<td>4.5</td>
<td>3.5</td>
<td>92</td>
</tr>
<tr>
<td>4</td>
<td>2.6</td>
<td>5.5</td>
<td>91.9</td>
</tr>
<tr>
<td>5</td>
<td>63</td>
<td>13</td>
<td>24</td>
</tr>
<tr>
<td>6</td>
<td>5</td>
<td>90</td>
<td>5</td>
</tr>
<tr>
<td>7.1</td>
<td>30</td>
<td>53</td>
<td>17</td>
</tr>
<tr>
<td>7.2</td>
<td>72</td>
<td>21</td>
<td>7</td>
</tr>
<tr>
<td>7.3</td>
<td>30</td>
<td>53</td>
<td>17</td>
</tr>
<tr>
<td>7.5</td>
<td>2.3</td>
<td>25.2</td>
<td>72.5</td>
</tr>
<tr>
<td>8</td>
<td>29.1</td>
<td>33.5</td>
<td>37.4</td>
</tr>
<tr>
<td>9</td>
<td>19.7</td>
<td>51.3</td>
<td>29</td>
</tr>
<tr>
<td>10</td>
<td>0</td>
<td>95</td>
<td>5</td>
</tr>
</tbody>
</table>

*SO$_4$, Na and minerals.

**Table 4: PM$_{2.5}$ speciation factors for each SNAP sector.**

<table>
<thead>
<tr>
<th>SNAP sector</th>
<th>BC [%]</th>
<th>POA [%]</th>
<th>Other* [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>7.4</td>
<td>2.9</td>
<td>89.7</td>
</tr>
<tr>
<td>2</td>
<td>16.5</td>
<td>73.9</td>
<td>9.6</td>
</tr>
<tr>
<td>3</td>
<td>2.7</td>
<td>4.3</td>
<td>93</td>
</tr>
<tr>
<td>4</td>
<td>3.4</td>
<td>5.3</td>
<td>91.4</td>
</tr>
<tr>
<td>5</td>
<td>63</td>
<td>13</td>
<td>24</td>
</tr>
<tr>
<td>6</td>
<td>5</td>
<td>90</td>
<td>5</td>
</tr>
<tr>
<td>7.1</td>
<td>30</td>
<td>53</td>
<td>17</td>
</tr>
<tr>
<td>7.2</td>
<td>72</td>
<td>21</td>
<td>7</td>
</tr>
<tr>
<td>7.3</td>
<td>30</td>
<td>53</td>
<td>17</td>
</tr>
<tr>
<td>7.5</td>
<td>1</td>
<td>23</td>
<td>75.9</td>
</tr>
<tr>
<td>8</td>
<td>39.4</td>
<td>47.9</td>
<td>12.7</td>
</tr>
<tr>
<td>9</td>
<td>22.2</td>
<td>61.4</td>
<td>16.4</td>
</tr>
<tr>
<td>10</td>
<td>0</td>
<td>90</td>
<td>10</td>
</tr>
</tbody>
</table>

*SO$_4$, Na and minerals.
No particulate matter is emitted in SNAP sector 7.4 which corresponds to non-exhaust (volatilization) road transport emissions. BC and POA contributions are low in SNAP sectors 1, 3 and 4 (less than 10%).

3.4. CHIMERE chemistry-transport model

The 3D CTM CHIMERE (Vautard, 2001; Schmidt, 2001) has been developed since 1997 by IPSL (including LISA) and INERIS. It was originally designed to provide (i) short-term predictions of ozone and aerosol concentrations and (ii) long-term (several years) predictions associated to emissions control scenarios. As a multi-scale model, it can simulate air quality in domains from hundreds (local scale) to several thousands of kilometers (regional scale), with horizontal resolution approximately from one to one hundred kilometers. CHIMERE thus allows operational air quality forecasting and simulations, and is used by different local air quality agencies (AASQA) in France and abroad. It is also used by the French organisation INERIS for daily air quality forecasting in France and Europe (PREV’AIR service http://www.prevair.org).

CHIMERE simulates transport, gas-phase chemistry, some aqueous-phase reactions, size dependent aerosol species including secondary organic aerosol formation, dry and wet deposition. Input data — meteorology, anthropogenic and biogenic emissions, boundary conditions, initial conditions, land-use — are prepared by various modules. It computes concentrations over a regular horizontal grid with hybrid sigma-pressure vertical discretization. Different gas phase chemical mechanisms are available, MELCHIOR1 (adapted from the EMEP mechanism) which describes more than 300 reactions of 80 gaseous species, or its reduced version MELCHIOR2 that includes 120 reactions of about 40 species. Considering seven species (primary particulate material, nitrate, sulfate, ammonium, biogenic secondary organic aerosol, anthropogenic secondary organic aerosol, water), distributed on eight size bins, CHIMERE treats coagulation, absorption, nucleation aerosol processes, as well as some multiphase chemistry. Inorganic aerosol thermodynamic equilibrium is calculated using the ISORROPIA model.

Boundary layer diffusion is represented using a vertical diffusivity depending on the surface friction velocity and the boundary layer height following Troen and Mahrt (1986). Boundary layer height can be diagnosed in CHIMERE from vertical profiles of meteorological parameters (temperature, wind, humidity). The boundary layer height is an input parameter given by the MM5 model. Vertical wind is also diagnosed through a bottom-up mass balance scheme. The numerical time solver is the TWOSTEP method.

3.5. Input data, model configuration and domains

Table 5 presents information the CHIMERE input data and its sources.

<table>
<thead>
<tr>
<th>Input data</th>
<th>Sources</th>
</tr>
</thead>
<tbody>
<tr>
<td>Meteorology</td>
<td>MM5 model</td>
</tr>
<tr>
<td>Boundary and initial conditions</td>
<td>INCA for gas species, GOCART for aerosols</td>
</tr>
<tr>
<td></td>
<td>(global climate models)</td>
</tr>
<tr>
<td>Land-use data</td>
<td>GLCF global land-use database</td>
</tr>
</tbody>
</table>

Table 5: Description of input data sources

Simulations are performed during the summer MEGAPOLI campaign (July 2009) with a five-day spin-up period. One-way nesting is used with two domains, CONT3 and MEG3. Table 6 gives

---

5 The spin-up period designates a period during which simulated concentrations can still be influenced by initial conditions. Basically, it corresponds to the time required to evacuate initialized air masses outside the domain: the larger the domain is, the longer spin-up period should be.

6 The one-way nesting consists in using concentrations simulated over a coarse domain as initial conditions in a refined more little domain.
details about this set-up.

**Table 6: Description of domains.**

<table>
<thead>
<tr>
<th>Domain name</th>
<th>Number of cells (lon x lat)</th>
<th>Spatial resolution (lon x lat)</th>
<th>Min longitude ; Min latitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>CONT3</td>
<td>67 x 46</td>
<td>0,5° x 0,5°</td>
<td>[-10,5° ; 35°]</td>
</tr>
<tr>
<td>MEG3</td>
<td>120 x 120</td>
<td>0,04° x 0,027°</td>
<td>[-0,35° ; 47,45°]</td>
</tr>
</tbody>
</table>

The domain is subdivided into eight vertical layers, from ground to more than 5000m height, with vertical resolution decreasing with altitude. The first three layers have a depth of about 40, 70 and 110 m, respectively. The model configuration is summarized in Table 7.

**Table 7: Description of model configuration.**

<table>
<thead>
<tr>
<th>Chemical scheme</th>
<th>Reduced MELCHIOR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sea salts</td>
<td>Considered inert</td>
</tr>
<tr>
<td>SOA chemistry scheme</td>
<td>Complex scheme</td>
</tr>
<tr>
<td>Advection scheme</td>
<td>First order Van Leer scheme</td>
</tr>
</tbody>
</table>

3.6. Simulated cases

Three simulations were performed with the two TNO inventories, without and with megacities refined data, respectively designated as MEGAPOLI and MEGAPOLI-MC (**Mega**Cities), and a third one designated as EMEP-LA (**Laboratoire d’Aérologie**, Toulouse, France). Table 8 gives the spatial resolution, the emitted species and their corresponding data sources.

**Table 8: Description of the three simulated inventories.**  
*Color code remains the same in all Figures and Tables.*

<table>
<thead>
<tr>
<th>Case name</th>
<th>Anthropogenic emitted species</th>
<th>Data source inventories</th>
<th>Longitude-latitude resolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>EMEP-LA</td>
<td>BCfin POAfin</td>
<td>LA/OMP (<a href="http://www.obsmp.fr">Observatoire Midi-Pyrénées</a>) inventory (C. Liousses, B. Guillaume, R. Rosset)</td>
<td>0.5x0.5°</td>
</tr>
<tr>
<td></td>
<td>SO₂ NOₓ NH₃ CO NMVOC CH₄ PMcoa</td>
<td>EMEP-MSC-west inventory</td>
<td>50km x 50km</td>
</tr>
<tr>
<td>MEGAPOLI</td>
<td>SO₂ NOₓ NH₃ CO NMVOC</td>
<td>First TNO emission inventory (Cf. MEGAPOLI deliverable 1.2) with TNO speciation factors, and <strong>without</strong> refined data for megacities</td>
<td>0.125x0.0625°</td>
</tr>
</tbody>
</table>

---

1 EMEP-LA corresponds to the default inventory in CHIMERE model.
In the CHIMERE model, only three particulate matter types — POA, BC and a so-called PPM representing mineral particulate emissions of anthropogenic origin — and three size classes — big for big particulate with sizes above 10 μm, coa for coarse particulate with sizes between 2.5 and 10 μm, and fin for fine particulate with sizes below 2.5 μm — can be considered in anthropogenic emissions.

In our study, we do not consider any big PM emissions. In the EMEP-LA case, particulate matter emissions in the fine mode only include POA and BC emissions, while only PPM is considered in the coarse mode. Particulate matter emissions in the MEGAPOLI and MEGAPOLI-MC cases are more detailed since POA, BC and PPM emission data are available in both coarse and fine modes, through the PM speciation factors.

### 3.7. Procedure to Compute Emission Factors

Figures 1 and 2 present the aircraft trajectory and the corresponding measured and simulated BC concentration time series for the 9th of July, 2009. Simulated concentrations are obtained by a 3D-space and time interpolation along the flight. In both observations and simulations, the Paris region plume is well distinguishable against background, and peaks can thus be located on the trajectory, giving the approximate central line of the plume. We focus in this study on the time period during which the flight altitude is rather constant (about 600 meters above ground) to avoid potential problems caused by not well established vertical mixing.

*Figure 1*: Observed (black line) and simulated (purple, blue and blue sky lines for EMEP-LA, MEGAPOLI and MEGAPOLI-MC inventories respectively) BC concentration time series during the 2009 July 9th ATR-42 flight; measured aircraft altitude time series (grey line). Note: Vertical lines indicate the plume peaks (corresponding to points in Figure 2).
Differences of plume shape between observations and simulations are mainly due to errors in simulated winds. Errors in emission inventories spatial structures can also lead to discrepancies in plume location, particularly if the inventory resolution is low (which is the case for the EMEP-LA inventory, compared to the MEGAPOLI and MEGAPOLI-MC inventories). The methodology used to get emissions correction factors can be sensitive to such errors; this point will be discussed later.

For both observations and simulations, the plume integrals of tracer concentration have to be calculated along the flight path perpendicular to the plume, and the background has to be subtracted. The background is computed as the 25 percentile of the time series. Peak integrals for successive transects are summed up. The ratio of peak area sums from simulations and observations gives for each flight an averaged emission evaluation factor for the Paris region (i.e. a factor above unity means overestimated emissions). Comparing simulated and observed peak areas instead of comparing maximum peak concentrations also avoids errors in the lateral dispersion of the plume.

It is not appropriate to compute all the area under the curve above the constant background because: (i) the observed background is sometimes noisy, (ii) there may exist some little peaks outside the Paris plume which need to be ignored since our analysis focuses on the emissions of the Paris region. The method used to isolate properly the plume is based on the assumption that its lateral dispersion is Gaussian, which is theoretically predicted for plumes from point sources. A Gaussian fit is applied to concentration time series around each peak, allowing us to deduce a time interval on which concentration integration would theoretically include the major part of the plume transect (±5σ). Figure 3 illustrates this Gaussian fitting procedure. It also shows that the approximation of a Gaussian lateral dispersion is reasonable. Integrals are consequently computed for each peak individually, and then combined for a particular day. The ratio of modelled area to measured area gives the emission correction factor for each peak and for the whole day.
Measured concentrations sometimes show short spikes (but including more than one value) presenting increases inside the plume at levels inconsistent with the general plume level. This can be due to local heterogeneities, or instrumental problems. In such cases, the curve is smoothed in order to erase such values.

Plume direction discrepancies between observations and simulations imply differences between the plume centerline and the airplane trajectory. This leads to differences in the apparent plume width and in the integral over the plume transect. This effect is corrected by applying multiplicative correction factor of $1/\sin(\text{angle})$ to the peak areas. Corrections are in general in the range 0-20%.

4. Results and Discussions

4.1. Emission evaluation factors

Figures 4 and 5 give the emission factors resulting from our analysis procedure, for each flight and each peak, and for BC and NO$_x$ respectively.

The 28$^{\text{th}}$ of July was removed because of too large errors in the plume position, leading to inconsistencies in the methodology. The figures show high day-to-day variability for all inventories and both species. Some days give particularly specific results, as on the 21$^{\text{st}}$ of July for BC and NOx where factors are very low, and the 13$^{\text{th}}$ of July for NOx where factors are quite high. Table 9 gives some statistical results over all flights, considering errors as multiplicative.

The results indicate significant differences between the three inventories for BC emissions with a potential overestimation for EMEP-LA and MEGAPOLI and an underestimation for MEGAPOLI-MC. The EMEP-LA and MEGAPOLI BC inventories are expected to be positively biased over the
Paris region compared to MEGAPOLI-MC, because of the population proxy used to distribute emissions in top-down inventories. Indeed, this proxy is suspected to not be fully relevant over big cities where, for example, biomass burning emissions per capita are assumed to be lower than in rural or suburban areas. The comparison suggests that the three inventories may be overestimating NOx emissions by 40%.

**Figure 4:** BC angle-corrected emission evaluation factors (ratio simulation over observation) for each flight and each peak simulated (observations in black, always equal to 1, EMEP-LA in purple, MEGAPOLI in blue and MEGAPOLI-MC in blue sky). **All** refers to the resulted emission factor for the whole flight including all the peaks.

**Figure 5:** NOy angle-corrected emission factors (ratio simulation over observation) for each flight and each peak (observations in black, always equal to 1, EMEP-LA in purple, MEGAPOLI in blue and MEGAPOLI-MC in blue sky). **All** refers to the resulted emission factor for the whole flight including all the peaks.
Table 9: Mean and uncertainty over all July flights for each inventory and each species.

<table>
<thead>
<tr>
<th>Inventory</th>
<th>BC</th>
<th>NOx</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean</td>
<td>One-sigma uncertainty factor</td>
</tr>
<tr>
<td>EMEP-LA</td>
<td>1.30</td>
<td>1.68</td>
</tr>
<tr>
<td>MEGAPOLI</td>
<td>1.19</td>
<td>1.66</td>
</tr>
<tr>
<td>MEGAPOLI-MC</td>
<td>0.74</td>
<td>1.61</td>
</tr>
</tbody>
</table>

However, uncertainties are strong for all inventories and both species, with a factor of about 1.6 leading to quite large one-sigma confidence intervals. Specific patterns on some days (13th and 21st of July) contribute significantly to this uncertainty. Thus, specific over or underestimations are not significant at the one-sigma confidence level (i.e. 1σ uncertainty ranges extending below and above unity). This prevents from concluding about a systematic under- or overestimation of inventories at this stage of the analysis.

4.2. Benefits and uncertainties of the methodology

The methodology presented in this Deliverable to evaluate NOx and BC emission inventories over Paris region intends to minimize several error sources: (i) modelled chemistry errors by considering inert tracer species, (ii) lateral dispersion and plume direction errors by considering integrated concentrations against concentrations, (iii) wind field errors by integrating over all peaks for a given flight. Based on aircraft measurements, the approach is assumed to be more relevant in terms of representativeness than comparisons with ground measurements, always suspected to be affected by a local situation with strong concentration gradients.

However, there are still several potential error sources that influence the plume structure and concentrations: (i) the meteorological input data, particularly PBL height and wind field, (ii) vertical mixing in the CHIMERE model, (iii) deposition and (iv) BC and NOy measurements, and finally (v) spatial and/or temporal pattern of emission inventories coupled to errors in wind fields.

The noticed strong day-to-day variability in emission correction factors can be due to emission variability, to the above mentioned uncertainty sources or to both. To evaluate the methodology, these various potential sources of variability have to be investigated.

4.3. Mixing layer depth adjustment

The mixing layer (ML) depth directly influences concentrations in the well-mixed plume and has therefore a strong impact on the calculation of emission factors. Continuous ML depth measurements are available at two sites in the Paris region: at SIRTA (background periurban site) and LHVP (background urban site, in Paris). From these observations, a simple adjustment is proposed to a posteriori correct possible errors in the model.

An overestimation in CHIMERE induces a stronger vertical dilution of the plume and lower concentrations. The approach adopted here is based on the assumption that vertical mixing is established (this point will be discussed further). In such cases, concentrations in the ML are proportional to the maximum ML depth reached since the beginning of the day. Nevertheless, vertical mixing needs some time to be effective after a ML depth change, roughly an hour. A decreasing boundary layer height creates a residual layer, and does not impact concentrations at sufficient distance from the emission sources. Our approach takes into account increasing ML depths, and assumes constant height in the case of decreasing ML. Then, relative differences between measurements and observations are calculated for each hour, giving an adjustment factor to apply to each peak area, according
to its occurrence time.

Figure 6: PBL height in meters at SIRTA and LHVP sites for the 9th of July. Crosses and points indicate hourly averaged observed PBL heights after the moving median fit applied, without and with the non-decreasing procedure respectively. Dotted and continuous lines indicate the simulated PBL height (in black for MM5 model, in blue for WRF model), without and with the non-decreasing procedure respectively.

Table 10: Mean ML depth relative error [in %] for each peak in each flight.

<table>
<thead>
<tr>
<th>Date</th>
<th>Peak 1</th>
<th>Peak 2</th>
<th>Peak 3</th>
<th>Peak 4</th>
<th>Peak 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>01/07/2009</td>
<td>−7.1</td>
<td>−7.1</td>
<td>−7.1</td>
<td>−8.7</td>
<td>−8.7</td>
</tr>
<tr>
<td>09/07/2009</td>
<td>15.6</td>
<td>11.6</td>
<td>11.6</td>
<td>5.3</td>
<td>5.3</td>
</tr>
<tr>
<td>10/07/2009</td>
<td>−14.2</td>
<td>−14.2</td>
<td>−19.9</td>
<td>−19.9</td>
<td>−</td>
</tr>
<tr>
<td>13/07/2009</td>
<td>6.8</td>
<td>13.4</td>
<td>13.4</td>
<td>12.1</td>
<td>12.1</td>
</tr>
<tr>
<td>15/07/2009</td>
<td>−11.1</td>
<td>−11.1</td>
<td>−11.1</td>
<td>−2.0</td>
<td>−10.7</td>
</tr>
<tr>
<td>16/07/2009</td>
<td>−11.2</td>
<td>−11.2</td>
<td>−18.3</td>
<td>−18.3</td>
<td>−22.9</td>
</tr>
<tr>
<td>20/07/2009</td>
<td>1.5</td>
<td>1.5</td>
<td>0.8</td>
<td>−1.2</td>
<td>−1.2</td>
</tr>
<tr>
<td>21/07/2009</td>
<td>−13.5</td>
<td>−0.9</td>
<td>−0.9</td>
<td>6.4</td>
<td>17.2</td>
</tr>
<tr>
<td>25/07/2009</td>
<td>−27.6</td>
<td>−27.6</td>
<td>−20.0</td>
<td>−20.0</td>
<td>−</td>
</tr>
<tr>
<td>28/07/2009</td>
<td>−6.0</td>
<td>−6.0</td>
<td>−7.1</td>
<td>−</td>
<td>−</td>
</tr>
</tbody>
</table>

Measured and modelled ML depths are sometimes particularly noisy seriously affecting our results. A moving median over a window of an hour is thus calculated to smooth the curve. Such a time scale is consistent with the fact that vertical mixing is not instantaneous and needs about an hour to be effective. Figure 6 illustrates the procedure.

As ML depth measurements are available at two sites, we consider the mean of both computed factors. Table 10 gives an overview of these mean adjustment factors for each peak in each flight.

Table 11 gives results obtained with that ML depth adjustment. All mean emission correction factors have decreased; illustrating the fact that ML depth tends to be underestimated by the model in July, as illustrated by July statistics given in Table 12.
However, results show no significant changes in the uncertainty factors which remain quite large. Therefore, despite their non-negligible importance, ML depth errors do not appear as a significant uncertainty source in emission evaluation factors.

**Table 11:** Mean emission correction factors and associated uncertainty over all July flights for each inventory and each species with (and without, previous results) ML depth adjustment.

<table>
<thead>
<tr>
<th>Inventory</th>
<th>BC with ML depth adjustment (without adjustment)</th>
<th>NOx with ML depth adjustment (without adjustment)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean One-sigma uncertainty factor One-sigma confidence interval</td>
<td>Mean One-sigma uncertainty factor One-sigma confidence interval</td>
</tr>
<tr>
<td>EMEP-LA</td>
<td>1.22 (1.30) 1.62 (1.68) 0.75 – 1.98 (0.78 – 2.18)</td>
<td>1.38 (1.46) 1.63 (1.63) 0.85 – 2.25 (0.90 – 2.38)</td>
</tr>
<tr>
<td>MEGAPOLI</td>
<td>1.12 (1.19) 1.61 (1.66) 0.69 – 1.80 (0.72 – 1.97)</td>
<td>1.26 (1.33) 1.78 (1.71) 0.71 – 2.24 (0.78 – 2.29)</td>
</tr>
<tr>
<td>MEGAPOLI-MC</td>
<td>0.69 (0.74) 1.59 (1.61) 0.44 – 1.10 (0.46 – 1.19)</td>
<td>1.34 (1.42) 1.81 (1.75) 0.74 – 2.42 (0.81 – 2.49)</td>
</tr>
</tbody>
</table>

**Table 12:** Statistical results of observed vs simulated ML depth at both SIRTA and LHVP sites, in the early morning (0-6 UTC) and afternoon (13-17 UTC). Units: meters.

<table>
<thead>
<tr>
<th>SIRTA</th>
<th>LHVP</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>0-6 UTC</td>
<td>13-17 UTC</td>
</tr>
<tr>
<td>RMSE</td>
<td>Bias</td>
</tr>
</tbody>
</table>

Assuming a simple inverse proportionality between plume concentrations and ML depth ignores the deposition sink which makes the considered species NOy and BC not fully conservative. Moreover, the vertical turbulent diffusion in CHIMERE model depends, among other parameters, on ML depth, and errors on that last variable can therefore lead to wrong vertical mixing state. Without ML depth measurements over (or at least closer to) the aircraft trajectory, investigating the ML depth at the two sites can be a crude approximation in case of important ML depth horizontal heterogeneities at the scale of a few hundred of kilometers, and therefore can lead to potential discrepancies.

**4.4. Case of the 21st of July**

As noticed before, some days give very different results compared to the average ones. For example, the 21st of July showed a strong underestimation of both BC and NOx emissions. An important question in this study is the reliability of our methodology during days with important errors. We investigate here the 21st of July case.

Firstly, Figure 7 shows quite high plume direction discrepancies during this day due to a wind direction error of roughly 20°. This is among the largest discrepancies encountered in this study.

Figure 8 shows wind speed measurements and simulations aboard the aircraft on the 21st of July. As illustrated by the statistical results given in Table 13, bias and RMS error on the 21st of July are the largest among all flights (1.77 m/s and 2.46 m²/s², respectively).

Such an important wind speed overestimation implies that we do not sample the same air masses in terms of age and emission contribution.
Figure 7: ATR-42 aircraft trajectory, the 21st of July, 2009 (black line). Observed (black points) et simulated (purple, blue and blue sky points for EMEP-LA, MEGAPOLI and MEGAPOLI-MC inventories respectively) Paris region plume position, retrieved by locating plume peaks on BC concentration time series.

Figure 8: Airborne measured (black line) vs MM5 (purple line) simulated wind speed during the 21st of July flight.

To illustrate more quantitatively this point, we consider the morning emission peak which occurs around 5 UTC. A 1.77 m/s bias implies that the 5 UTC emissions can travel an additional distance of 38 km after six hours (11 UTC, beginning of the constant altitude flight), and 57 km after nine hours (14 UTC, end of the constant altitude flight), than for the case of a correct wind. This is to be compared to the farthest distance reached by flights which is about 150 km from Paris. Moreover, the 21st of July is one of the windiest days, with roughly 10 m/s of wind at about 600 m above ground. At this speed, 216 and 324 km can be covered by air masses respectively in six and nine hours. Thus, contrary to the other days, when in general morning emissions are sampled during the flight path, emissions occurring around noon are sampled by the aircraft in this case.

About ML depth, SIRTA and LHVP sites give opposite results with our previous adjustment procedure, as illustrated in Figure 9.

The model underestimates ML in SIRTA and overestimates it in LHVP. This incoherence is mainly due to very high values measured at SIRTA in the morning, after 7 UTC, where ML depth directly reaches about 2300 m before it decreases to about 1800 m. For these different reasons (largest error
in wind speed, non-representative wind speed, inconsistency in ML height corrections), we decided to remove July 21 from the rest of the analysis.

**Table 13:** Statistical results of airborne measured vs MM5 simulated wind speed for all the July flight.

<table>
<thead>
<tr>
<th>Date</th>
<th>Bias (m/s)</th>
<th>RMSE (m²/s²)</th>
<th>Correlation</th>
</tr>
</thead>
<tbody>
<tr>
<td>01/07/2009</td>
<td>−0.68</td>
<td>1.16</td>
<td>0.185</td>
</tr>
<tr>
<td>09/07/2009</td>
<td>−0.64</td>
<td>1.63</td>
<td>0.131</td>
</tr>
<tr>
<td>10/07/2009</td>
<td>0.16</td>
<td>1.29</td>
<td>−0.019</td>
</tr>
<tr>
<td>13/07/2009</td>
<td>0.07</td>
<td>1.64</td>
<td>−0.095</td>
</tr>
<tr>
<td>15/07/2009</td>
<td>0.15</td>
<td>2.00</td>
<td>−0.064</td>
</tr>
<tr>
<td>16/07/2009</td>
<td>0.02</td>
<td>1.35</td>
<td>0.143</td>
</tr>
<tr>
<td>20/07/2009</td>
<td>−0.24</td>
<td>1.46</td>
<td>0.018</td>
</tr>
<tr>
<td><strong>21/07/2009</strong></td>
<td><strong>1.77</strong></td>
<td><strong>2.46</strong></td>
<td><strong>0.433</strong></td>
</tr>
<tr>
<td>25/07/2009</td>
<td>0.35</td>
<td>1.32</td>
<td>0.517</td>
</tr>
<tr>
<td>28/07/2009</td>
<td>−1.28</td>
<td>2.17</td>
<td>−0.056</td>
</tr>
<tr>
<td><strong>All flights</strong></td>
<td><strong>0.00</strong></td>
<td><strong>1.71</strong></td>
<td><strong>0.701</strong></td>
</tr>
</tbody>
</table>

**Figure 9:** PBL height in meters at SIRTA and LHVP sites the 21st of July. Crosses and points indicate hourly averaged observed PBL heights after the moving median fit applied, without and with the non-decreasing procedure respectively. Dotted and continuous lines indicate the simulated PBL height (in black for MM5 model, in blue for WRF model, not used in our study), without and with the non-decreasing procedure respectively.

New results are given in Table 14. All mean emission correction factors increase and, more important, uncertainty factors significantly decrease.

The EMEP-LA inventory one-sigma confidence interval now extends above one, suggesting a potential overestimation of BC and NOx emissions. Uncertainties are still too large in our analysis to conclude on a systematic error (always on the one sigma confidence level).
Table 14: Mean emission evaluation factors and associated uncertainties for each inventory and each species with ML depth adjustment, over all July flights except the 21st of July flight (and with that day included, previous results).

<table>
<thead>
<tr>
<th>Inventory</th>
<th>Mean</th>
<th>One-sigma uncertainty factor</th>
<th>One-sigma confidence interval</th>
<th>Mean</th>
<th>One-sigma uncertainty factor</th>
<th>One-sigma confidence interval</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>EMEP-LA</td>
<td>1.39 (1.22)</td>
<td>1.33 (1.62)</td>
<td>1.05 – 1.85 (0.75 – 1.98)</td>
<td>1.57 (1.38)</td>
<td>1.42 (1.63)</td>
<td>1.11 – 2.23 (0.85 – 2.25)</td>
</tr>
<tr>
<td>MEGAPOLI</td>
<td>1.27 (1.12)</td>
<td>1.35 (1.61)</td>
<td>0.94 – 1.72 (0.69 – 1.80)</td>
<td>1.44 (1.26)</td>
<td>1.59 (1.78)</td>
<td>0.90 – 2.29 (0.71 – 2.24)</td>
</tr>
<tr>
<td>MEGAPOLI-MC</td>
<td>0.77 (0.69)</td>
<td>1.42 (1.59)</td>
<td>0.55 – 1.10 (0.44 – 1.10)</td>
<td>1.55 (1.34)</td>
<td>1.59 (1.81)</td>
<td>0.97 – 2.46 (0.74 – 2.42)</td>
</tr>
</tbody>
</table>

4.5. Hourly emission contribution to plume

Concentrations measured in the plume at a given time integrate emissions from different times from various locations, in the Paris region. As an example, to illustrate that point, we emitted and transported tracers within the CIHMERE model each hour at a particular point in the centre of Paris. By comparing tracer concentrations from different hours at the incidence of the emission peak, the contribution of emissions from specific hours can be deduced, as illustrated in Figure 10. Each peak integrates emissions mainly over a 4-hours time period. If we consider all the flight, this time period can reach 7 hours.

This analysis points out a critical limitation in our methodology: we cannot discriminate emission spatial and temporal patterns; our method allows evaluation of emissions over a specific time period, depending on the wind speed, and over a specific geographical area. Therefore, we not only evaluate inventories, but also the emission diurnal (and monthly) profiles introduced in the CIHMERE emissions preprocessor. Indeed, inventories give annual emissions by species and by SNAP sector; temporal distributions by month, by type of day (week/weekend) and by hour then need to be applied.

Finally, we have here two uncertainty sources that are difficult to clearly evaluate: the spatial and temporal pattern of emissions, the first depending on the inventory and its resolution, the second depending on the input wind field data and temporal emission profiles used in CIHMERE.

As Paris region emissions do not a perfect circular symmetry, plume direction discrepancies can lead to an error in emission correction factors.
4.6. Deposition

BC and NO_y (particularly HNO_3) deposition directly affects concentrations in the plume. If CHIMERE overestimates deposition, concentrations in the plume as well as emission factors will consequently be underestimated. Gas dry deposition is usually represented as a process that is linearly dependent on concentrations at ground through a deposition velocity.

Precipitation measurements in the Paris region and MM5 meteorological model agree quite well in the spatio-temporal rainfall patterns, with rain mainly occurring on the 16th, and to a lesser extent on the 9th, 13th and 21st of July. If we focus on the aircraft trajectory and on the flying time period, we can confidently state that no important washout occurred during flights, which considerably limits wet deposition.

Dry deposition effects are more important, and an important error source, since no observations are available to constrain this process and the model shows quite important deposition rates. Figure 11 illustrates the dry deposition effects on mixing ratios in the plume. Almost 1 ppbv of NO_y is lost by dry deposition for the example of July 9, which roughly corresponds to 10-20% of its total concent-
trations. However, both the peak values and the background are affected, which tends to minimize the actual error on the peak area.

Considering the CHIMERE uncertainties for these processes of roughly a factor of two, their sensitivity on emission factor results is still need to be investigated.

4.7 Other errors

Measurement of absorption coefficients and BC content

The Particle Soot/Absorption Photometer (PSAP) is used to measure in near real time the light absorption coefficient. The method is based on the integrating plate technique in which the change in optical transmission of a filter caused by particle deposition on the filter is related to the light absorption coefficient of the deposited particles using Beers Law. During MEGAPOLI the 3 wavelength (467, 530 and 660 nm) version has been operated on board the ATR-42 research aircraft. We applied several corrections to the PSAP measurements to obtain absorption coefficients at the three wavelengths and to deduce the black carbon (BC) content from the absorption coefficient extrapolated to 637 nm, thus, comparable to BC derived from state of the art MAAP instruments.

The PSAP calibration and correction methods are described in detail in Bond et al. (1999), Virkkula et al. (2005), and Müller et al. (2010). Corrections of PSAP measurements require corrections for the PSAP spot size, for aerosol particle scattering (from simultaneous TSI nephelometer measurements) to take into account the lowering of transmittance of the filter due to scattering, for the absolute transmittance (filters have been changed after every flight, transmittance never decreased below 0.9), the PSAP flow had been calibrated as a function of upstream pressure.

To estimate the uncertainty of PSAP measurements following values reported by Bond et al. (1999) at 95% confidence: flow rate calibration, spot size correction (19.23 mm²), response to scattering (TSI nephelometer), and extrapolation to 637 nm (the MAAP wavelength) are typically a few percent, the response to absorption is roughly 22%, and the instrument precision (unit-to-unit variability) is smaller than 6%. The total estimated uncertainty of PSAP measurement at typical atmospheric absorption levels encountered during the MEGAPOLI measurement campaign is 15-20%. The absorption coefficient calculated at 637 nm was converted to a black carbon (BC) content using the specific absorption of 6.6 m²g⁻¹ also used internally in the MAAP instrument.

Another source of uncertainty might be the aircraft community aerosol inlet (CAI). This isokinetic and isoaxial CAI is based on the University of Hawaii shrouded solid diffuser inlet designed by A. Clarke and modified by Meteo France. The CAI inlet allows for entirely sampling the submicron particles and partly sampling of supermicron particles with an upper 50% sampling efficiency for supermicron particle sizes at diameter around 5 µm (McNaughton et al., 2007; Gomes et al., in preparation).

All these uncertainty sources may contribute to the random error or precision of the method, and manifest in the important day to day variability of the adjustment factors. They also can contribute to a systematic error, which has not been quantified up to now, given that the statistical uncertainty in general does not allow concluding on a systematic emission under or overestimation.
Conclusions

The evaluation of emission inventories is of great importance since, as an input for air quality models, their uncertainties are spread over many physical and chemical processes in chemistry transport models. This report has presented an original methodology to evaluate NOx and BC emission inventories over the Paris region. Three emission inventories have been evaluated, EMEP-LA, MEGAPOLI and MEGAPOLI-MC, the two latter using high resolution, and the last one integrating local emissions data from AIRPARIF. The second inventory has been specifically designed for the FP7 MEGAPOLI project. Based on airborne measurements, our approach is assumed to be more robust, as compared to the use of surface measurements with questionable spatial representativity. The MEGAPOLI summer campaign has made available a large dataset of ten daily flights in the Paris plume.

By considering quasi inert tracer species, BC and NOy, the approach is not seriously affected by errors related to atmospheric chemistry. By comparing time integrated concentrations and not only concentrations, it minimizes errors in lateral dispersion and small errors in plume direction. Various sources of uncertainty still remain in such a methodology. They have been identified and include boundary layer height estimates, vertical mixing, deposition and wind speed. For the boundary layer height, a simple adjustment procedure was developed to a posteriori correct for potential discrepancies between simulations and measurements. Some flights were removed because of large errors in wind speed and/or mixing layer depth. Other uncertainty sources were discussed but still need to be more deeply investigated.

Mean results suggest that inventories may overestimate BC and NOx emissions, except MEGAPOLI-MC which may underestimate BC emissions. However uncertainties of the estimated emission correction factors are quite large for all inventories and both species. Considering the one-sigma confidence interval, it appears that only the EMEP-LA inventory significantly overestimates BC and NOx emissions. For the two other inventories, uncertainties are still too large to clearly conclude on systematic under- or overestimation. However, as expected BC emissions from the MEGAPOLI cadastre are larger than those from MEGAPOLI-MC (with specific Paris information).

The availability of evaluated and corrected emissions inventories is of great importance for assessing pollution transformation processes within the MEGAPOLI project. The developed methodology is of general interest for evaluating megacities emissions.
Acknowledgements

The MEGAPOLI project is very thankful and acknowledges the Laboratoire d’Hygiène de Paris (LHVP), The SIRTA/IPSL, le Golf de la Poudrerie à Livry-Gargan for hosting campaign sites. Without this help, the campaign would not have been possible. Also University Paris-Est, Créteil, INRA/Gignon, Météo-France, Roissy and ENPC, Marne la Vallée are thanked to host lidar instruments. It is also grateful to all voluntary participants who made this campaign a great success. Yohann Morille, Jean-Charles Dupont and Patrick Chazette are thanked for the PBL heights and LIDAR wind speed data.

The research leading to these results has received funding from the European Union’s Seventh Framework Programme FP/2007-2011 within the project MEGAPOLI, grant agreement n°212520.

The PhD thesis (H. Petetin) is supported by an Ile-de-France DIM grant.

All contributions from the MEGAPOLI campaign team are acknowledged:

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Scientific Report 10-08, MEGAPOLI-11-REP-2010-03, 19p, ISBN: 978-87-993898-1-0
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