An attempt at estimating Paris area CO₂ emissions from atmospheric concentration measurements

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11 Abstract

12 Atmospheric concentration measurements are used to adjust the daily to monthly budget of fossil fuel CO₂ emissions of the Paris urban area from the prior estimates established by the 13 14 Airparif local air quality agency. Five atmospheric monitoring sites are available, including one at the top of the Eiffel tower. The atmospheric inversion is based on a Bayesian 15 16 approach, and relies on an atmospheric transport model with a spatial resolution of 2 km with boundary conditions from a global coarse grid transport model. The inversion adjusts a prior 17 18 knowledge about the anthropogenic and biogenic CO₂ fluxes from the Airparif inventory and 19 an ecosystem model, respectively, with corrections at a temporal resolution of 6 hours, while 20 keeping the spatial distribution from the emission inventory. These corrections are based on 21 assumptions regarding the temporal autocorrelation of prior emissions uncertainties within the 22 daily cycle, and from day to day.

The comparison of the measurements against the atmospheric transport simulation driven by the a-priori CO_2 surface fluxes show significant differences upwind of the Paris urban area, which suggests a large and uncertain contribution from distant sources and sinks to the CO_2 concentration variability. This contribution advocates the inversion should aim at minimizing model-data misfits in upwind-downwind gradients rather than misfits in mole fraction at individual sites. Another conclusion of the direct model-measurement comparison is that the CO_2 variability at the top of the Eiffel tower is large and poorly represented by the model for most wind speed and directions. The model inability to reproduce the CO_2 variability at the heart of the city makes such measurement ill-suited for the inversion. This and the need for constraining the budgets for the whole city suggests to assimilating upwind-downwind mole fraction gradient between sites at the edge of the urban area only.

5 The inversion significantly improves the agreement between measured and modelled 6 concentrations gradients. Realistic emissions are retrieved for two 30-day periods and suggest 7 a significant overestimate by the AirParif inventory. Similar inversions over longer periods 8 are necessary for a proper evaluation of the optimized CO_2 emissions against independent 9 data.

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11 **1. Introduction**

Although the total CO₂ emissions of developed countries may be well constrained from the 12 total consumption of fossil fuel, its spatial and temporal distribution are not known with the 13 same level of accuracy. In so-called bottom-up emission estimates, CO₂ emission is 14 15 calculated as a combination of geo-referenced activity proxies (e.g. road traffic data, or 16 number and type of buildings that relate to residential emissions, (Gurney et al., 2012)) 17 multiplied by emission factors, accounting for the disaggregation of national annual budgets 18 when dealing with regional or city inventories. The accuracy of the bottom-up inventories is 19 seldom assessed and mostly relies on the difference between various estimates and on expert 20 knowledge.

21 Due to the high population density associated with ground transportation, residence and 22 industry, anthropogenic CO₂ emissions are large within cities (Pataki et al., 2006). The 23 emitted CO₂ is transported in the atmosphere and results in elevated CO₂ concentration above and downwind of cities. There is therefore a potential to estimate the net CO_2 flux of a city 24 25 from a few atmospheric concentration measurements located within or in the vicinity of the 26 city (McKain et al., 2012). Over a very dense urban area, the net CO₂ flux is dominated by 27 fossil fuel emissions, but over less dense urban structures, the net ecosystem exchange (NEE) 28 becomes significant and can partly offset fossil CO₂ emissions during the growing season 29 (Nordbo et al., 2012). Top-down net CO₂ flux estimates, constrained by independent 30 atmospheric measurements, could come in complement to, or for the assessment of, current 31 estimates that rely on bottom-up inventories.

The technique of estimating surface CO₂ fluxes from atmospheric composition measurements 1 2 -and potentially from prior information- is relatively mature. It has been used for many years to estimate the biogenic fluxes at the global (Gurney et al., 2002; Chevallier et al., 2010), 3 4 continental (Broquet et al., 2013; Peylin et al., 2005) and regional (Lauvaux et al., 5 2009;Lauvaux et al., 2012) scales. However, because of uncertainties in the atmospheric transport, insufficient measurement sampling, and inconsistencies between the mathematical 6 7 framework hypothesis of most inversions (e.g. no biases, Gaussian distribution of errors, 8 uncorrelated observation errors) and the reality, the results are not always consistent, in 9 particular at the regional scale, as shown for instance through the recent comparison of global 10 and continental-scale biogenic flux estimates by several global inversons (Peylin et al., 2013).

Estimating the net CO₂ flux of a city amplifies using similar mathematical and modelling 11 The spatial 12 tools amplifies the difficulties inherent to the atmospheric inversion. 13 heterogeneity of the source and the possibility of having very high emissions locally (e.g. a power plant) make the structure of the prior error statistics complex and the concentration 14 plume highly variable. Relating mole fractions to city sources further requires accurate 15 16 atmospheric transport model at fine scale. Atmospheric transport in urban areas is influenced by specific meteorological processes such as higher roughness of urban canopies (Zhao et al. 17 2014) and urban heat island effects (Nehrkorn et al., 2013). For instance, (Pal et al., 2012) 18 19 reported significantly thicker boundary layer over the Paris city than in the surrounding rural 20 area during a four day campaign that took place in March 2011, which was interpreted as a 21 consequence of the urban heat island effect. Another difficulty, shared with the inversion of biogenic fluxes, lays in the temporal variability of the fossil fuel emissions, which have a 22 23 strong daily cycle but also day-to-day variability resulting from, for instance, temperature changes (through heating) or activity (e.g. traffic) variability. Last, measurements in and 24 around a target city collect CO₂ molecules of various origins that must be separated into city 25 26 sources and remote sources and sinks through the inversion.

This challenge has been addressed recently by several research projects, e.g. INFLUX (sites.psu.edu/influx, (Shepson et al., 2011)) over Indianapolis city or Megacities (<u>http://megacities.jpl.nasa.gov</u>; (Duren and Miller, 2012)) over Los Angeles, which have setup a network of surface, tower and airborne measurements of the atmospheric CO_2 mole fractions. Satellite data may also provide valuable information as shown by (Kort et al., 2012). The results from the on-going urban CO_2 measurement project at Salt Lake City indicated that monthly emission relative changes of 15% could be detected at the 95%
 confidence level with the current monitoring system (McKain et al., 2012) even though this
 study concluded on the inability to derive absolute estimates for a given month.

4 The CO₂-MegaParis project has a similar objective for the Paris area. This is a potentially 5 favourable case as the city is very dense and the emissions intense over a limited surface, with 6 a fairly flat topography in the surroundings, which makes the atmospheric transport modelling easier. A pilot campaign early 2010 was conducted in the framework of the MEGAPOLI 7 8 project. Measurements of the mole fraction of CO₂ and its isotopes have been used to 9 estimate the relative contribution of fossil and biogenic emissions in the concentration 10 gradients (Lopez et al., 2013). The main campaign started in August 2010 with the installation of three CO₂ and CO monitoring stations within the city and its surrounding that 11 provided near-continuous measurements until July 2011. These three stations complement 12 two stations of the ICOS France network located in the Paris region outside the city that have 13 14 been operational for several years. (Lac et al., 2013) made a first analysis of the measurements and a comparison against atmospheric modelling using the Meso-NH 15 16 mesoscale transport model, combined with a surface scheme that accounts for the urban environment, for a period of 5 days in March 2011. They demonstrated the ability of the 17 18 modelling framework to reproduce several features of the mixing layer height, as reported in 19 (Pal et al., 2012), and of the mole fraction daily cycle.

20 Large efforts have been made by AirParif, the air quality agency for the Paris area, to generate an inventory of the Paris area emissions, for various pollutants and for CO₂ as well. The 21 22 AirParif emission inventory, detailed in section 2.2, provides an hourly description of the CO₂ 23 emissions at ≈ 1 km resolution for representative weekdays and months. We use this 24 inventory as an input to the atmospheric transport simulations and compare the results to the atmospheric concentration measurements from the five sites. We then attempt a correction of 25 the inventory based on the differences between the observed and modelled mole fractions. 26 27 With only 5 stations in the vicinity of the city, there is likely not enough information to constrain the spatial distribution of the emissions. We therefore only rescale the emissions, 28 relying on the spatial distribution provided by the Airparif inventory. For the inversion, NEE 29 30 and fossil fuel emissions are optimized separately. We focus on two 30-day periods in the fall 31 of 2010. This choice is driven by the expectation of rather small biogenic fluxes during this 32 time period, which makes easier the interpretation of the measurements in terms of

anthropogenic fluxes. Our objective is to assess whether a reliable estimate of the emissions 1 2 at the daily to monthly time scales can be derived from the combination of atmospheric measurements, available inventories and information on the atmospheric transport. 3 Α 4 forthcoming paper will apply the methodology to a full year of observations and analyse the 5 result for the spring and summer periods, when CO₂ uptake by NEE can partially offset fossil 6 fuel emissions (Pataki et al., 2007). In the following, section 2 analyses the time series of 7 measured and modelled CO₂ mole fractions; section 3 describes the methodology to correct 8 the inventory based on the measurement-model mismatches. The results are shown in section 9 4 while section 5 discusses the results and concludes.

10 **2.** Measurements and direct simulations

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2.1. CO₂ concentration measurements

12 In this paper, we use CO₂ mole fraction measurements that have been acquired continuously 13 in the framework of the CO2-Megaparis and ICOS-France projects. Three stations have been 14 equipped with high precision CO₂/CO analysers (Picarro G1302) specifically for the project objectives. One is located in the heart of Paris, at the summit of the Eiffel tower, 300 m 15 16 above the surface. Two are located in the North and North-East of the Paris area in a mixed urban-rural environment. They are complemented by two ICOS-France stations that were 17 18 operational before the start of the project. One is located in the South-West, about 20 km 19 from the centre of Paris, while the other is a tall tower located further south by about 100 km. 20 Both use gas chromatograph analysers (Agilent HP6890). The location of the stations are given in Table 1 and shown in Figure 1. They are very roughly located along a NE-SW 21 22 direction, which defines the dominant wind directions, thus favourable for the monitoring of the CO₂ increase due to the emissions of the Paris area, with a station at the edge of the urban 23 24 area in both directions. The measurements are quality-controlled and binned at a temporal 25 resolution of 1 hour. They have been regularly calibrated against the WMO mole fraction scale (Zhao and Tans, 2006) so that measurement accuracy to the WMO-X2007 scale is 26 estimated to be better than 0.38 ppm. The instrumental reproducibility is better than 0.17 27 28 ppm on the 5 minute average measurements available from the CO2-Megaparis stations, and 29 the temporal averaging to the hourly-mean values used in this paper leads to precision much better than the accuracy (Zhao and Tans, 2006). 30

1 2.2. Atmospheric transport modelling

2 Atmospheric transport modelling provides the link between the surface fluxes and the atmospheric mole fractions. Here, we use the Chimere transport model (Menut et al., 2013) 3 4 with a resolution of 2 km around the Paris city, and 10 km for the surrounding of the modelling domain (see Figure 1). There are 118x118 pixels in the modelling grid that covers 5 an area of approximately $500 \times 500 \text{ km}^2$. There are 19 layers on the vertical, from the surface 6 7 to 500 hPa. The Chimere transport model is driven by ECMWF-analysed meteorology at 15 8 km resolution. There is no urban scheme in the atmospheric modelling that is used here, 9 which may be seen as a significant limitation to our inversion set-up. However, we conducted 10 forward simulation comparisons between our modelling and that used in (Lac et al., 2013), 11 which includes specific surface parameterization to account for the urban area, and we did not 12 find significant differences on the simulated CO₂ mole fractions.

13 The model simulates the mole fractions that are driven by the surface fluxes and the boundary 14 conditions. The surface fluxes that are accounted for in the simulations are the sum of

- Anthropogenic fossil fuel CO₂ emissions within the Île-de-France region, from the
 AirParif inventory, as described in section 2.3 and shown in Figure 2. Île-de-France is
 the administrative region spreading typically within 60 km around the Paris city, the
 boundaries of which are shown in Figure 1.
- Anthropogenic fossil fuel CO₂ emissions outside the Île-de-France region, according
 to the Edgar database [Edgar, 2011] available at 10 km resolution. These are only
 annual mean fluxes, and there is no description of the diurnal or seasonal cycle in this
 inventory.
- Biogenic fluxes from the C-TESSEL land surface model, as described in section 2.4.

The CO₂ boundary conditions prescribed at the lateral and top edges of the simulation 24 25 domain, and transported inside the domain by Chimere, are obtained from the Monitoring Atmospheric 26 Composition and Climate (MACC) global inversion, v10.2) 27 (http://www.copernicus-atmosphere.eu/). In this simulation, the global distribution of surface CO₂ fluxes has been optimized to fit the mole fractions measured at a number of stations 28 29 distributed over the world, given their assigned uncertainty and prior information of the 30 surface fluxes. Given the relatively coarse spatial resolution of the transport model used in the MACC inversion, CO₂ boundary conditions here are temporally and spatially very smooth
and have little impact on the spatial gradients simulated within the domain area.

3 **2.3. AirParif Inventory**

The AirParif air quality agency (http://www.airparif.asso.fr/en/index/index) has developed an inventory of emissions (for greenhouse gases such as CO₂ but also for air pollutants) at 1 km spatial resolution and hourly time step for the Île-de-France region. The emissions are quantified by activity sectors. The improvement of methodologies and emission factors lead to frequent updates of the emission estimates.

9 Nearly eighty different source types are included in the inventory with three main classes: 10 point sources, linear and diffuse sources. Point sources correspond to large industries, power 11 plants, and waste burning; linear sources are related to transportation, while diffuse sources 12 are mostly associated to the residential and commercial sectors. The road traffic emission 13 estimates use a traffic model and vehicles counting devices that report the number of vehicles 14 and their average speed over almost 40 000 km portions of roadways. Large industries are 15 requested to report their CO₂ emissions and these are used in the inventory. For smaller industrial sources that are not required to report their emissions, a disaggregation of the 16 17 regional fuel consumption is made based on the number of employees, leading to larger 18 uncertainties. We have used the latest available version of the inventory, corresponding to 19 year 2008, which has been developed for 5 typical months (January, April, July, August, and 20 October) and three typical days (weekday, Saturday and Sunday) to account for the seasonal 21 and weekly cycle of the emissions. Therefore this inventory estimates typical emissions but 22 does not attempt to reproduce the daily variations resulting from specific meteorological 23 conditions, or specific events such as public holidays.

Figure 2 shows an example of the spatial distribution of the total emissions for a weekday in October. Typical values are a few hundred $gCO_2 \text{ m}^{-2} \text{ day}^{-1}$ within the city and a few tens $gCO_2 \text{ m}^{-2} \text{ day}^{-1}$ in the suburbs. The main roads are clearly shown with flux enhancements of a few tens $gCO_2 \text{ m}^{-2} \text{ day}^{-1}$, at the 1 km² resolution of the inventory. Further processing of this map shows that one third of the Île-de-France emissions are within 10 km of the Paris centre, and 61% are within 20 km.

There is a large temporal variation of emissions, as shown in Figure 3, mostly at the daily scale, but also at the weekly and seasonal scales. Most components show a large daily cycle with minimum emissions at night. During the day, the traffic related emissions show several maxima, in the morning, midday, and late afternoon. The daily cycles of the other activities are less pronounced but nevertheless significant. Point sources have the smallest daily cycle amplitude due to the industrial temporal profile that is relatively flat. The Paris area has few point sources and they contribute to typically 20% of the total emissions. The seasonal cycle is most pronounced for the residential emissions related to heating and cooking. One notes that residential CO_2 emissions do not go to zero during the summer months, because energy is still consumed for cooking and for heating water in summer.

In the following, the AirParif inventory for year 2008 is used as a prior estimate of the fossil fuel emissions within the Île-de-France region, both for the direct transport simulations (section 2.5) and for the flux inversion (section 3). Note that the inventory of point source emissions provides injection heights that have been used in the source term of the simulations. The AirParif inventory is provided as a function of legal time, and we have accounted for the time shift between legal time and UTC time, including the impact of daylight saving. Note that, due to the longitude of Paris, UT time and solar times are very similar.

15 **2.4. Biogenic Fluxes**

16 The Net Ecosystem Exchange fluxes used here are provided by the land surface component of 17 the ECMWF forecasting system, C-TESSEL (Boussetta et al., 2013). They are extracted from 18 the ECMWF operational archives at the highest available resolution, 15 km and 3 hours. 19 These data are interpolated in space (2 to 10 km) and time (1 hour) to be consistent with our 20 atmospheric transport model grid and temporal resolution.

21 Figure 4 shows the mean daily cycle of NEE for the Île-de-France area and for the 12 calendar 22 months. There are large diurnal and seasonal NEE cycles. The flux is positive (emission) 23 during the night and negative (uptake) during the day, even during the winter months, given 24 the rather mild winter temperature prevailing over the Paris area. Nevertheless, the amplitude 25 of the daily cycle of NEE is much larger in summer than it is in winter. The NEE values are of similar magnitude than the anthropogenic emissions with a strong anti-correlation on the 26 daily cycle (negative NEE vs. large anthropogenic emissions during daytime; positive NEE 27 and smaller anthropogenic emissions during the night). During the winter, NEE is relatively 28 small and the anthropogenic emissions clearly dominate, but daytime NEE still offsets on 29 average $\sim 20\%$ of the emissions, according to the C-TESSEL model simulations. During 30 31 spring and summer, however, the daytime NEE uptake is larger in absolute value than the 32 anthropogenic emissions as shown through a comparison of Figures 3 and 4.

As our main interest is the anthropogenic emissions, we chose to analyse a period when the biogenic flux is small, i.e. during fall and winter. The present paper focuses on two 30-day periods that start on October 21st and November 27th 2010. During these periods, the monthly mean hourly NEE fluxes are less than 3 ktCO₂ per hour over the Île-de-France area. NEE is then small, but not negligible, compared to anthropogenic emissions during the chosen inversion periods.

7 **2.5. Direct CO₂ transport simulations**

8 Figure 5, together with Figure S-1 in the supplementary, shows the time series of the CO_2 9 mole fractions together with an indication of the modelled wind speed and direction to help 10 the interpretation of the results. These time series are derived from observations and direct 11 atmospheric modelling as described in section 2.2.

12 The Trainou (TRN) station (bottom row) is far from the Paris agglomeration. In addition, the 13 measurement inlet is at 180 m from the surface. It shows a diurnal cycle amplitude that is much smaller than at the other sites. In addition, the modelled contribution from both 14 15 anthropogenic and biogenic fluxes within the simulation domains is limited to a few ppm, as shown by the difference between the black and green curve. There are a few exceptions 16 17 however, essentially when the wind blows from the North, i.e. from the Paris city direction, and transports fossil CO₂ from the urban area to the TRN rural site. The best examples are 18 around Dec 8th and Dec 23rd. For these particular cases, the measurements at TRN are 19 significantly larger than the model results. The underestimate by the model is not limited to 20 21 these dates and there are significant discrepancies between the model and the measurements at 22 this remote background site, in particular at the end of November and at the beginning of 23 December.

24 The other sites are much closer to Paris and are then more affected by the fossil CO₂ 25 emissions. At Gif-sur-Yvette (GIF) the largest mole fractions are observed when the wind is from the North-East, which is expected as the Paris city is in that direction. There is also an 26 27 impact of the wind, as the largest mole fractions are measured in low wind speed conditions. During the Oct-Nov period (Figure S-1), the wind is mostly from the South and South-West, 28 29 thus not from the city, and there is a relatively good agreement between the modelled and 30 measured mole fractions. In December, the wind direction is more variable, the fossil CO₂ 31 signal appears much larger, and there are very significant differences between the 32 measurements and the model estimates.

Gonesse (GON) is located to the North of the city, while Montgé-en-Goële (MON) is further 1 2 away to the North-East. The shorter distance to the main source may explain the larger signal at the former station. The only cases when the modelled anthropogenic contribution is small 3 4 at GON (small difference between black and green curve) is when the wind is from the North. 5 For other wind directions, the modelled signal is strong -more than 10 ppm- and there are 6 large differences between the measurements and the modelling results. During December, the 7 measurements are most often larger than the model estimates. A similar observation can be 8 made at MON. Surprisingly, the measurements are significantly larger than the modelling 9 results, even when the wind blows from the North or North-East, i.e. when the Paris agglomeration contribution is negligible (Dec 3rd, Dec 6-9, Dec 22-23). For these cases, the 10 most likely explanation is an underestimate of modelled CO₂ from the boundary conditions or 11 from emissions within the modelling domain outside of Île-de-France. Hereafter, we shall 12 13 denote this contribution as that from "remote fluxes". Note that this impact from remote 14 fluxes shows a large increase of the mole fraction for the periods discussed above. We may then hypothesize that this increase is underestimated. The interpretation is that anthropogenic 15 16 emissions from the Benelux area generate high concentrations that are underestimated in the 17 boundary condition field that is used in our simulations.

18 The EIF site is at the top of the Eiffel tower, 300 m above the Paris city. The wind speed for 19 this station is larger than for the other one, simply because it is higher in altitude. One expects 20 atmospheric mixing between the surface emissions and the inlet, so that the measurements are 21 representative of a larger area than e.g. MON and GON. Nevertheless there are some very significant differences between the modelled and the observed mole fractions at EIF. The 22 differences may be huge, larger than 30 ppm, even during the afternoon, e.g on Oct 24th, Nov 23 7th, Dec 3rd, Dec 12th. Clearly, our atmospheric modelling framework cannot properly 24 25 represent the mole fraction time series at the EIF station, either because of strong local (sub 26 grid cell) emissions, or because of atmospheric transport processes that are not properly 27 represented, in particular concerning the vertical transport above the city. Further analysis of 28 the model-measurement mismatch is shown in Figure S-3. The largest mismatches are 29 preferentially observed during the morning and for low wind speeds, but are observed at all hours of the day and for all wind speed and directions which prevents from attributing these 30 mismatches to a specific bias in the transport model or to a bias in the estimate of the 31 emissions for a specific area. 32

The curves in Figure 5 and Figure S-1 show very large temporal variations of CO₂ within a 1 2 day at all stations. Further analysis confirms that the largest variations are observed during the night, when the mixing layer is shallow. During the night and morning, the atmosphere is 3 4 often very stable so that surface emissions accumulate within the lowest atmospheric layers, 5 the thickness of which ranges from a few meters to tens of meters. The atmospheric mole 6 fraction is then mostly sensitive to local fluxes and vertical mixing -an atmospheric process 7 that is difficult to model- so that there is a large uncertainty about the modelled link between 8 the emissions and the atmospheric mole fraction. The night-time and morning measurements 9 are thus not appropriate for our flux inversion, as inverting them would be too sensitive to 10 atmospheric transport biases. As a consequence, we focus on the concentration measurements 11 acquired during the afternoon only, from noon to 4 p.m., when the mixing layer is usually 12 well developed. The daily averages of these afternoon measured and modelled values are 13 shown in Figure 5 as diamond symbols.

14 **2.6.** Analyses and insight for the inverse modelling configuration

Both the measurements and the modelling results show some impact of the Paris area anthropogenic emissions on the CO_2 mole fractions at the 5 sites analysed here. The mole fraction increases over the modelled large-scale value depends on the wind speed and direction and a typical order of magnitude is 10 ppm. As expected, the signal is smaller for the rural station of TRN, which is further away from the city than the other sites. Many of the features in the measured time series are well reproduced by the modelling framework, which gives some confidence in its usefulness to improve the emission estimates.

There are also some significant differences between the measured and modelled mole 22 23 fractions that cannot be justified by inaccurate emission inventories in the Paris area. The most obvious such feature is the mole fraction underestimate at MON and GON in northerly 24 25 wind conditions when these sites are little sensitive to the Île-de-France emissions. This 26 feature strongly suggests that remote fluxes lead to mole fraction increases that have biases 27 with a typical magnitude that is similar to the impact of the Paris area emissions. On the other 28 hand, as the impact from remote fluxes is large scale, one may expect that this impact is 29 similar for monitoring stations upwind and downwind from the Paris urban area. The model-30 measurement error may then be strongly reduced when analysing the difference of mole fractions between two stations that are located upwind and downwind the Paris urban area, 31 32 respectively. On the other hand, the mole fraction difference between such stations that are

1 close to the Paris area should contain a clear signature of the emissions from this area, and a 2 relatively weak signature from other fluxes. It then suggests the use of downwind-upwind 3 gradients in the CO_2 mole fractions rather than the absolute value of CO_2 measurements in the 4 inversion procedure.

5 The other significant feature in the comparison of the modelled and measured CO₂ mole 6 fractions is much larger errors at the EIF site than at the other stations. These results illustrate 7 the difficulty in modelling the CO₂ mole fraction within cities, even with a measurement inlet 8 in altitude, well above the sources. Note that (McKain et al., 2012) also find very large (>30 9 ppm) model-measurement mismatches within the urban area of Salt Lake City, even when 10 using a high-resolution model. Similarly (Lac et al., 2013) finds large model-measurements differences at EIF despite the use of an urban parameterization in the modelling. The inability 11 to properly model the CO₂ signal at EIF may have detrimental impact on the emission 12 estimates derived from atmospheric inversion. Conversely, the forward simulations show that 13 14 the TRN site is little sensitive to the Paris area emissions due to its location further away from the city than the other sites. Consequently, it cannot be used as a "downwind" site; in 15 addition, GIF is better suited as an "upwind site" for southerly conditions as it is closer to the 16 17 urban area and provides therefore a better information on the air composition as it enters the 18 city. These features suggest not to use EIF and TRN and rather focus on MON, GON and 19 GIF to estimate the Paris area emission from their measured mole fractions.

20 The main objective of the "gradient" inversion method is thus to focus on the monitoring 21 stations that are at the edge of the urban area and to estimate the city scale emissions by 22 removing most of the upwind signal from the measured and modelled concentrations. The 23 upwind signal is driven by remote fluxes both from the boundary conditions and by fluxes 24 within the model domain but outside the city whose estimates bear very large uncertainties. The inversion method also attempts to select the downwind measurements that are affected by 25 the emissions from a large part of the city, in an attempt to minimize the impact of 26 aggregation errors. Ideally, we would select only the wind direction when one station lies 27 directly downwind from another, with the Paris city in between. However, given the very 28 29 limited network of stations surrounding Paris, we have to broaden significantly the range of 30 acceptable wind directions.

Based on this analysis, the emission estimate procedure only uses the measurements from GON, MON and GIF and is based on the CO_2 mole fraction gradients between the upwind

1 and downwind stations, a method which requires the selection of favourable wind conditions.

2 The mathematical framework is described in the next section while the inversion results are3 presented in section 4.

4 **3.** Flux inversion

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3.1. Principles

We follow a linear Bayesian inversion approach with Gaussian error statistics to determine the optimal surface fluxes (anthropogenic emissions and biogenic fluxes) and their uncertainties from a prior estimate of the fluxes and their uncertainties and from the mole fraction measurements.

We call **x** the state vector that gathers the scaling factors for the 6-hourly flux maps, $\mathbf{x}_{\mathbf{B}}$ its prior estimate, **H** the matrix operator that relates state parameters and mole fraction gradients according to the atmospheric transport model, **y** the observed mole fractions gradients, $\mathbf{y}_{\mathbf{F}}$ the simulated impact on these mole fraction gradients of the lateral boundary conditions and of the fluxes that are not accounted for in the state vector, **B** the uncertainty covariance matrix of $\mathbf{x}_{\mathbf{B}}$, and **R** the error covariance matrix of **y**. These components are detailed in the next section. The optimal solution is given by (Tarantola, 2005):

17 $\mathbf{x}_{\mathbf{A}} = \mathbf{x}_{\mathbf{B}} + (\mathbf{B}^{-1} + \mathbf{H}^{\mathrm{T}} \mathbf{R}^{-1} \mathbf{H})^{-1} \mathbf{H}^{\mathrm{T}} \mathbf{R}^{-1} (\mathbf{y} - \mathbf{y}_{\mathrm{F}} - \mathbf{H} \mathbf{x}_{\mathrm{B}})$ (1)

18 and its posterior error covariance matrix is

 $\mathbf{A} = (\mathbf{B}^{-1} + \mathbf{H}^{\mathrm{T}} \, \mathbf{R}^{-1} \, \mathbf{H})^{-1} \tag{2}$

Note that A does not depend on the actual measurement values, but varies, among other
factors, with their temporal and spatial sampling.

22 **3.2. State vector: x**

Both the anthropogenic and biogenic prior fluxes described in Section 2 show a large diurnal cycle that impacts the model simulations of CO_2 , and that is uncertain. It then appears useful to invert this cycle together with the flux daily mean values. However, as discussed earlier, only CO_2 measurements during the early afternoon can reliably be used to estimate the fluxes and their information about the daily cycle is rather poor. We limit the number of independent periods to 4 corresponding to the local times between 0-6 h, 6-12 h, 12-18 h, and 18-24 h, respectively.

For the fossil fluxes, we use a scaling factor for each individual day in the state vector, which 1 2 makes the number of corresponding variables amount to 30×4=120 for the 30-day period of the inversion. These scaling factors apply to the prior flux estimates derived from the AirParif 3 inventory and are noted λ_{0-6}^i , λ_{6-12}^i , λ_{12-18}^i , λ_{18-24}^i with i between 1 and 30. 4

5 Similarly, we optimize scaling factors of the prior NEE flux from C-TESSEL. The simulation 6 domain shown in Figure 1 is split into 3×3 large boxes, and we choose the same 6-hour periods than for the anthropogenic fluxes to optimize scaling factors of NEE. However, we 7 do not attempt a daily retrieval of NEE, and considered a single scaling factor for optimizing 8 9 monthly NEE each 6-hour window over a 30-day inversion period. The number of variables to optimize NEE is therefore $3 \times 3 \times 4 = 36$. In the following, these NEE scaling factors are 10 shown as α_{0-6}^X , α_{6-12}^X , α_{12-18}^X , α_{618-24}^X where X is one of the 9 large boxes. One of the 9 11 12 boxes covers the Île-de-France region while the other ones are in the surrounding. In the Inversion results sections, we analyse the inversion of NEE for the centre box (X=C) together 13 14 with those for the anthropogenic emissions. The surrounding boxes provide some ability to 15 the inversion system to control part of the errors from remote NEE, but one cannot expect to get reliable estimate of the NEE in these areas given the weak observational constraint on this 16 17 remote NEE.

The state vector **x** for the linear inversion has therefore 120+36 = 156 variables that represent 18 19 the scaling factors to the modelled fluxes. The prior value of each of these scaling factors in 20 $\mathbf{x}_{\mathbf{B}}$ is 1.

21

3.3. Measurements gradients: y

y contains the measurements gradients that are used to constrain the flux inversion. As 22 23 explained above, we only use hourly measurements that have been acquired during the 24 afternoon from noon to 4 p.m. local time. In addition, the corresponding measurements need 25 to have a sensitivity to local, unresolved, fluxes that is insignificant in comparison to that of 26 larger scale fluxes. This condition is not met when the wind speed is low. We therefore use 27 for the inversion only the measurements filtered for wind speeds larger than a given threshold 28 at both sites used to compute the gradient. The results presented in this study are obtained with a threshold of 2 m s^{-1} . The wind speed estimate used for such a selection is the one 29 30 analysed by the ECMWF at the location, height, and time of the observation. This criterion 31 retains about 70% of the potential measurements.

In Equation (1) the downwind-upwind differences in mole fraction measurements \mathbf{y} are corrected for the contributions that are not accounted for in the state vector (\mathbf{y}_F) . \mathbf{y}_F are the modelled mole fraction accounting for the boundary conditions and anthropogenic fluxes outside Île-de-France (prescribed from the Edgar database). This contribution is shown as a blue line in Figure 5 and Figure S-1.

6 When the wind is from the South-West (upwind direction between 160° and 260°), GIF is 7 considered as upwind from the urban area, and the corresponding y elements are the 8 differences between the mole fractions measured at either MON or GON and that measured at 9 GIF. Similarly, when the wind is from the North-East (upwind direction between 0 and 10 135°), MON is used as an upwind reference to the GIF or GON mole fraction measurements. 11 For other wind directions, the measurements are not assimilated.

12 **3.4. Prior flux uncertainties and error correlations: B**

Although we invert the scaling factors of fossil CO₂ emissions for each day and each 6-hour period, the uncertainties in these factors are correlated. We therefore attempt to assign correlations for the prior uncertainties based on several considerations: (i) the monthly budget for the AirParif inventory is generally stated to have an uncertainty of 20% which is used here; (ii) we assume small positive correlations between the different 6-hour windows; (iii) we assume stronger correlations from day to day for a given 6-hour window; (iv) the a priori uncertainty of individual 6-hour emission should have a typical order of 50%.

20 Based on these considerations, we set, rather arbitrarily, prior error correlations to 0.4 for two 21 adjacent time periods (e.g. 12-18 and 18-24) and to 0.2 for non-adjacent time period (e.g. 6-12 22 and 18-24). For successive days, we use an exponential de-correlation with a characteristic time T_{cor} . The correlation between the prior uncertainties of the fossil CO₂ emissions scaling 23 24 factors is then the product of this exponential and the time-periods correlation. For instance, the correlation between λ_{0-6}^5 and λ_{6-12}^9 is 0.4 exp $\left(-4/T_{cor}\right)$. The results shown in this paper 25 have been mostly obtained with a temporal correlation T_{cor} of 7 days, but other values, from 1 26 27 to 30 days, have been also tested. We have verified that such a **B** matrix is positive-definite. The desegregation of the assumed 20% uncertainty for the monthly emission totals, based on 28 these temporal correlations, results in a standard deviation of uncertainties for individual 6-29 30 hour period of 33% (T_{cor} =30 days) to 50% (T_{cor} =7 days).

1 For the biogenic flux scaling factors, we set a relative prior uncertainty (standard deviation) 2 close to 0.70 with some variations according to the box size (the variance varies inversely to the surface of the box), based on the numbers derived at 0.5° resolution in (Broquet et al., 3 2011). We do not assign any spatial / temporal correlation between the various biogenic 4 5 scaling factors, i.e. between the 9 boxes or the 4 time periods. Similarly, there is no correlation in **B** between the prior uncertainties on the biogenic and anthropogenic fluxes. 6

7 3.5. Operator matrix: H

8 The operator matrix **H** provides the link between the surface fluxes and the mole fraction 9 measurements. It combines the spatio-temporal distributions of the fluxes, both for the 10 AirParif inventory and the C-Tessel biogenic fluxes, that are assumed and not modified 11 through the inversion, the atmospheric transport by the Chimere model, the sampling of the 12 atmosphere at the instrument locations and the selection of gradients according to the criteria 13 developed in section 3.3. Note that the AirParif inventory has a 1 hour temporal resolution. 14 The direct simulation $(\mathbf{H} \mathbf{x})$ uses the description of the emissions at this temporal resolution. 15 Each element of the state vector corresponds to a natural or anthropogenic surface flux for a larger time period. We use the atmospheric transport model to compute the impact to the 16 17 mole fraction of each surface flux (156 in total) corresponding to an element of the control vector. The 4D mole fraction fields from each of these simulations are then sampled at the 18 place and time of the atmospheric observations used to compute the downwind-upwind 19 20 gradients corresponding to the observation vector. These simulated mole fraction gradients 21 provides the elements of each column of the H matrix.

22

3.6. Observation error: R

23 The measurements provided by the instrument are precise, certainly better than 0.3 ppm. 24 However, the observation error in **R** also includes any source of misfit between the model and 25 the data that is not accounted for in the state vector such as the representation error, the 26 impact of the error in the spatial distribution of the fluxes, and the atmospheric transport 27 modelling error. These are difficult to assess (Broquet et al., 2013) although one expects 28 significant values given the very heterogeneous urban environment that is discussed here.

29 Due to the complexity and misunderstanding of the processes underlying the observation error, that may lead to positive or negative correlations, we ignore observation error 30 correlations in the construction of **R**, which is thus diagonal. 31

We use two statistical diagnostics of the misfits in the observation space described by (Desroziers et al., 2005) to infer typical observation error variances: (i) the agreement between the sum of the uncertainty from the prior estimate of the control parameters and of the observation error with the RMS of the prior misfits to the assimilated data; and (ii) the agreement between the observation error with the mean of the product of prior and posterior misfits to the assimilated data. Based on this analysis, we set a 3 ppm observation error for the mole fraction gradients that are used for the inversion.

8 We can note that this value is significantly smaller than the model-measurement differences 9 as shown in Figure 5. This is due to the fact that the observation errors related to uncertainties 10 in the large scale impact of the remote fluxes are strongly correlated between the 11 measurement sites at a given time. Therefore, they vanish when considering gradients in the 12 model fractions rather than values at individual sites such as in Figure 5. This is further 13 discussed in section 4.2.

14 **4. Inversion results**

In the following, we present the result of the inversion described in the previous section. We first analyse the modelled mole fractions, prior and posterior, against the measurements. We then analyse the retrieved fluxes, both NEE and fossil fuel.

18

4.1. Mole fraction gradients

19 Figure 7 and Figure S4 show the time series of the afternoon-mean mole fraction gradients. 20 Some days are missing either because either station is unavailable or because the wind 21 direction does not fulfil the selection criteria developed in section 3.3. The prior value is 22 almost always positive, because the reference is chosen upwind the Paris agglomeration. There are a few exceptions, like on Dec 22nd at GON, MON being used at the upwind 23 24 reference according to the wind direction. As GON is in the northern part of the Paris 25 agglomeration, one expects a smaller signal than for southerly wind conditions. Further 26 investigation demonstrated that this unexpected behaviour is linked to a large spatial gradient of the CO₂ concentration generated by anthropogenic emissions over the Benelux accounted 27 for in the Edgar inventory and transported by the Chimere model (y_F in equation 1). 28 29 Interestingly, the observations confirm the sign and the order of magnitude of the gradient 30 that is modelled with our setup that uses crude anthropogenic emissions outside Île-de-France.

1 Another negative gradient is observed at GIF-MON for northerly wind conditions on Dec 3rd.

2 This is very unexpected and we could not find a valid explanation for this particular case.

In general, the observations are smaller than the prior, and the posterior is in between.
Indeed, the inversion result leads to concentration gradient that are closer to the observations.
As a result, some of the posterior gradients are negative (see end of the period at GIF in
Figure 7).

7 Figure 8 and Figure S4 show scatter plots of measured versus modelled mole fractions 8 gradients. The first row of the plots on each of these figures shows the modelled mole 9 fractions from the domain boundaries and the fossil CO₂ emission outside Île-de-France (black lines in Figure 5, y_F in equation 1) against the measurement. This constitutes the 10 11 modelled contribution to the mole fraction that is not optimized by the inversion. The values 12 on the Y-axis show the modelled impact of the remote fluxes on the upwind-downwind mole 13 fraction gradient. As expected, this impact is small compared to the measured gradient shown 14 on the X-axis.

15 The second row shows simulated CO₂ induced by prior NEE and fossil CO₂ fluxes (i.e. those 16 that are optimized through the inversion) against measured mole fractions corrected for the 17 large scale values (i.e. y_F , shown on the Y-axis of the first row). Although there is a large 18 spread, the correlation is significant, which shows that the transport model and the prior flux 19 set up have altogether some ability to reproduce the observed CO₂ mole fraction variability. 20 For the Oct-Nov period (in supplementary), the biases are large for all site gradients (2.1 to 4.8 ppm) whereas, for the Nov-Dec period, they are even larger at GIF-MON (7.1 ppm) but 21 22 rather small in comparison at both other sites. The standard deviation of the measurement-23 model difference varies with the sites and period, between 2.0 and 5.8 ppm. This is 24 significantly smaller than the standard deviation for the mole fractions (Figures 6 and Figure S2) that vary between 3.6 and 6.6 ppm. These smaller values confirm the choice made of 25 26 attempting an inversion based on the mole fraction gradient rather than the individual 27 observations.

After the inversion, the agreement is significantly improved as shown in the third row. Note however that the standard deviation for the MON site (when GIF is used as a reference) is slightly degraded from the prior value of 2.0 ppm. After the inversion, the correlation between optimized and observed CO_2 gradients for all three stations is larger than 0.90. For the other time period shown in the supplementary material (Figure S-5), the correlation statistics are not as good. However, this is due to a lower variability of the gradients, and the posterior standard deviations are 2.3, 2.7 and 2.3 ppm for the three sites, and are then similar as the values shown in Figure 7.

4 Overall, the statistics improve significantly between the prior and the posterior, and there is a 5 good agreement between the measured and modelled mole fraction gradients. This raises 6 confidence in our ability to model the impact of the Paris CO_2 emissions on the atmospheric 7 concentrations for various wind conditions.

8 **4.2. Daily flux estimates**

4.2. Daily hux estimates

9 Figure 9 shows the daily anthropogenic fluxes inferred by the inversion procedure. Here, we 10 have aggregated the 4 6-hour periods as well as their uncertainty, accounting for the error 11 correlations between the periods. Although the inversion controls scaling factors, we show 12 here the resulting fluxes expressed in MtCO₂ per day. There is a clear weekly cycle on the prior emissions that are smaller during the week-ends. One may also note a shift in prior 13 emission between Oct 29th and Nov 1st that corresponds to a change of month and therefore 14 the switch to a different dataset in the AirParif inventory. The Airparif inventory includes a 15 profile for October. For November and December, Airparif recommends the use of the 16 17 January emission profile.

18 The uncertainty reduction is significant for all the days of the two time periods and a typical 19 order of magnitude is a factor of 2. The emission uncertainty is reduced even for days with 20 no usable measurements, when the wind direction is not within any of the two ranges defined in section 3.3, due to the temporal correlation of the uncertainties and thus of the corrections 21 22 applied to the prior (section 3.4). The deviations of the flux estimate from the prior follow the 23 gradient observation deviation from the model (see Figure 7). These deviations are mostly 24 negative, although they are positive for a few days during both time periods. For the Nov-25 Dec period, the posterior emission estimates are within the bounds of the prior uncertainty range. On the other hand, the posterior estimate is much lower than the prior flux during the 26 second half of the Oct-Nov period (Figure 9, top). Interestingly this period (Nov 1st to Nov 27 28 20st, 2010) was very mild [Meteo France, 2010] which suggests that the heating sector emissions were well below the AirParif inventory values for that period. During this season, 29 30 according to the AirParif inventory, the heating sector, commercial and residential, amounts to more than 50% of the emission, so that the total emission is highly sensitive to temperature. 31 32 Note that AirParif recommends the use of the January inventory for both November and

December. As the temperatures are generally milder during October than January, one may
 expect that the inventory is larger than the true fluxes during October, which is then consistent
 with the negative correction to the fluxes during that period.

Figure 9 was generated using a 7 day correlation time for the emission uncertainties. We also 4 5 tested similar inversions using different error correlation times (T_{cor}) in the range of the 6 synoptic to seasonal time scales that drives the emission variability to assess the result sensitivity to this parameter. With a 1 day error correlation time, rather than 7 days used in 7 8 our standard configuration, there are days with little or no flux constrain by the observations, 9 while there is no smoothing of the day-to-day variability correction, resulting in an even 10 larger spread of the retrieved fluxes (not shown). At the other extreme, a 30-day correlation time leads to much smoother results. Most of the daily-optimized flux estimates remain within 11 the prior uncertainty range. 12

13 **4.3. Monthly budgets**

Figure 10 shows the monthly mean flux estimates for the Île-de-France region for the various 6-hour periods. It shows the results of the inversion for the anthropogenic emissions, the NEE of the central box that covers Ile de France, as well as the total. Note that the total estimate is necessarily the sum of the biogenic and anthropogenic fluxes. Conversely, the uncertainty range of the total is not a simple sum as it accounts for the correlations between NEE and fossil CO_2 emission errors in the **A** matrix linked to the difficulty in distinguishing NEE and fossil fluxes from the measurements.

The inversion has little impact on the fluxes for the 0-6h and 18-24h periods. On the other hand, the impact is strong for the 6-12h and 12-18h periods. This is because we only use afternoon observations that are sensitive to the emissions from the morning and afternoon periods only. The assigned correlations in the setup of the B matrix transport some constrain to the other time windows. Although the inversion based on the mole fraction gradients uses few independent observation, because of the additional data selection based on the wind direction, the impact on the flux estimates is significant.

Figure 10 shows that the uncertainty reduction is much larger for the fossil fuel than for the NEE. This is the result of the inversion based on the gradient downwind-upwind from the city which are mostly sensitive to the fluxes in between. The contribution from the NEE to the measurement is then small. Nevertheless, the correlations on the anthropogenic and NEE uncertainties are small (±0.15 or less) These numbers indicate that the observation sampling provides significant information to distinguish NEE from fossil CO₂ fluxes in the inversion. Although a given measurement cannot trace the origin of the mole fraction excess, the assigned biogenic and anthropogenic flux errors have different spatial and temporal patterns which are exploited by the inversion system to attribute the mole fraction signal to specific sectors. However, this attribution relies on the a-priori spatial and temporal distribution of the fluxes that are affected by uncertainties. Thus, the theoretical ability of the system to disentangle natural and anthropogenic fluxes may not be realized in practice.

8 5. Discussion and Conclusions

9 This paper is a first attempt at estimating the Paris area emissions from measurements of 10 atmospheric CO₂ mole fractions and prior flux knowledge. There is obviously room for 11 improvement in several aspects of the inversion system: the number and spatial distribution of 12 the monitoring stations, the atmospheric transport model including the use of an urban 13 scheme, the modelling of concentration at the simulation domain boundaries, the definition of the emissions outside Île-de-France, the definition of the control vector, etc. However, first 14 15 conclusions of broad implications beyond this first attempt can be drawn, that should guide 16 further inverse modelling developments for Paris and other cities.

17

18 The analysis of the CO₂ time series shows significant differences between the measured and modelled mole fractions upwind the Paris city. These differences indicate that the simulated 19 20 mole fraction at the domain boundaries may be off by several ppm. The errors in this simulation is of similar magnitude as the signal from the Paris area emissions. Although the 21 22 number of cases is limited, it seems that the boundary concentrations are significantly 23 underestimated when the wind is from the North or North-East (Benelux). These 24 uncertainties on the domain boundaries generate large scale errors in the modelled mole 25 fraction and suggest applying the inversion not on the measurements themselves, but rather on upwind-downwind gradients as was done in this paper. Indeed, the measurement-model 26 27 agreement is much better for the gradients than it is for the direct values. It confirms that the large-scale pattern of CO_2 mole fraction, which is not related to the Île-de-France fluxes, is 28 not properly modelled. The information provided by our five-site network does not allow 29 30 optimizing the structure of the CO₂ boundary conditions, which is directly prescribed by a 31 coarse scale global inversion. Exploiting the distant sites currently operational in Europe 32 would unlikely improve this situation. In this context, the inversion based upon gradients as

presented in section 4 appears necessary. It relies on the assumption that, due to atmospheric 1 2 diffusion, the signature of remote fluxes upwind the city is sufficiently homogeneous in space, horizontally and vertically, and time over the path through the city from upwind to 3 downwind sites both located within the afternoon PBL. As a consequence, the main part of 4 5 such a large-scale signal is removed through the differences between two sites. The validity 6 of this hypothesis is confirmed by the much better agreement between measured and 7 modelled mole fractions as shown through the comparison of Figure 6 and Figure 8. Both measurements and atmospheric transport simulations indicate, however, that the CO₂ mole 8 9 fraction signal generated by distant sources outside the Chimere model domain has some 10 spatial structures (see e.g. the variability of modelled values in Figure 8-top) which needs to 11 be accounted for.

The drawback of using the gradient-based inversion method is a reduction in the number of observations, in particular with the current monitoring network that only samples a fraction of possible wind directions. Nevertheless, although the number of observations is very much reduced, our inversion system based on the gradient reports significant uncertainty reductions. It must also be noted that we assumed a 7-day error correlation time for the anthropogenic emissions, so that our system shows flux uncertainty reductions, even on days with no valid observation as the flux is constrained by observation of the previous or following days.

19 The setting of temporal error correlation on prior fluxes is therefore essential for the 20 inversion. Although the results in this paper are mostly derived with a 7-day correlation 21 length, this is a somewhat arbitrary choice, and the results are significantly affected when 22 using different values. In particular, a much shorter value (1 day) leads to very large 23 variations in the posterior daily emissions. Further work should be devoted to the assignment 24 of objective correlation lengths based on the processes that lead to emission uncertainties. 25 Climatic conditions in general, and more specifically temperature during the cold season, influence the emission with a time scale that is consistent with synoptic events, i.e. close to a 26 week; the impact of specific events such as holidays, commemorations or strikes have a much 27 shorter time scale, while inventory biases linked to e.g. the emission factors have an impact 28 29 on the fluxes on time scales of months or even larger.

30

31 Our analysis also indicates model-measurement discrepancies at the EIF site that are much 32 larger than at other sites. On the one hand, this is somewhat surprising as measurement inlet

1 in altitude should insure a larger spatial representativeness than at the surface sites and less 2 sensitivity to local, poorly represented, emissions. Usually, tall tower-based measurements are preferred to those at the surface for the estimate of biogenic fluxes. On the other hand, 3 4 EIF is located close to the centre of the Paris city and is therefore affected by stronger local 5 emissions than the other sites used in this paper. City fluxes are highly heterogeneous while 6 the model used in this paper has a 2 km spatial resolution, does not include information on the 7 3D structure of the urban canopy, and uses limited information on the CO₂ source injection 8 heights. Such model may then be insufficient to properly account for atmospheric processes 9 that link the local surface fluxes to the concentrations at the top of the Eiffel tower. Previous 10 results obtained at MeteoFrance by (Lac et al., 2013) using a high (2 km) resolution 11 meteorological model that includes urban parameterizations, and validated against local 12 meteorological measurements, also show high model-data misfits at EIF, similar to those 13 found in the present paper. (McKain et al., 2012) also show a poor skill at representing the 14 mole fraction at urban sites, so that the information content of the measurements is not applied for an estimate of the absolute emissions, but rather for a on long term relative 15 change. These findings can be related to our difficulties for modelling urban CO₂ at EIF using 16 a 2 km resolution transport model are typical of the current generation of models. The use of 17 18 urban sites such as EIF for atmospheric inversion will likely necessitate long term research by 19 the inverse modelling and transport modelling communities.

20

21 At present, our mesoscale atmospheric transport model cannot reconcile the measurements at 22 the top of the tower with those at the surface in the vicinity of the city, given our set of 23 surface fluxes and inversion settings. This cast doubts on the quality of the modelling at the 24 other sites. Indeed, if the atmospheric transport model does not properly simulate the 25 atmospheric vertical transport between the surface and an inlet at 300 m in altitude, it likely misrepresents the link between surface fluxes and atmospheric mole fractions. Conversely, 26 27 the large modelling errors at EIF may be related to its urban location (and to the strong influence of local urban sources) and this would raise concerns regarding the ability to exploit 28 29 urban measurements, and therefore to solve for the spatial distribution of the fluxes within the 30 urban area.

The largest differences between the measured and modelled concentrations occur for low 1 2 wind speeds. For this reason, we have chosen a 2 m s⁻¹ wind speed threshold below which the measurements are not used in the inversion. A larger threshold rejects further observations, 3 4 and reduces the range of flux corrections through the inversion. The choice of the threshold is 5 somewhat arbitrary and we have refrained from using a large one to clearly demonstrate the 6 impact of a few situations with low wind-speed. There are several hypotheses for the poor 7 modelling at low wind speed, including larger representativity errors of subgrid patterns, or larger errors in vertical mixing modelling. However, such issues are continuous and there is 8 no indication that the modelling errors disappear between e.g. 2 and 3 m s⁻¹. Thus, further 9 rejection of low wind-speed observations may hide the deficiencies in the atmospheric 10 11 transport without improving the flux inversion.

12

13 We also stress that our analysis is based on measurements during the late fall period. This is a favourable case for the inversion of fossil fuel CO₂ emissions as there is less interference with 14 15 the biogenic fluxes (Pataki et al., 2007). During spring and summer, the NEE is much larger 16 (in absolute value) and also more uncertain. In fact, during May, the biogenic sink is likely larger than the anthropogenic emissions within Île-de-France as shown by Figure 3 and Figure 17 18 S4. The gradient inversion method is designed to also minimize this interference of biogenic 19 flux with the constraint on anthropogenic fluxes. Indeed, the theoretical posterior 20 uncertainties indicate little correlations between the retrieved NEE and anthropogenic 21 emissions. There is however vegetation within the urban area that may generate a significant 22 sink during the growing season. A successful anthropogenic emission inversion would benefit from additional efforts for describing the biogenic fluxes and the use of additional 23 tracers such as ¹⁴C to separate the signature of fossil fluxes and biogenic emissions. One 24 future direction is thus to use a more realistic NEE model over the Paris area, that could be 25 26 calibrated upon local eddy covariance observations (e.g. the method used in (Gerbig et al., 2003)) and satellite land cover and vegetation activity. 27

28

The prior estimate of the Île-de-France CO_2 emissions does not account for the human respiration. Yet, within dense urban areas, human respiration can be a significant fraction of the fossil fuel emissions (Ciais et al., 2007) (Widory and Javoy, 2003). Respiration by human beings is a source of CO_2 of typically 1 kg CO_2 day⁻¹ (Prairie and Duarte, 2007) which,

assuming a total population of 11.7 millions for the Île-de-France, leads to 4.2 MtCO₂ per 1 year, or 8% of the AirParif fossil fuel inventory. Although small, this flux is far from 2 negligible compared to fossil fuel emissions. While the CO₂ mole fraction measurements are 3 sensitive to the human respiration flux, our control vector only accounts for the fossil fuel 4 5 emissions and NEE fluxes. Although it does not have point sources, the spatial distribution of 6 the human respiration is broadly similar to that of the fossil fuel emissions, so that the 7 inversion will attribute the human respiration mole fraction signal to the fossil fuel rather than 8 the NEE fluxes. We therefore expect an overestimate of the fossil fuel emission by typically 9 8% in our inversion that neglects human respiration. A larger percentage may be expected in 10 summer and a smaller in winter due to the seasonal cycle of the fossil fuel emissions that has 11 a larger relative amplitude than that of the human respiration. Improvement of our inversion system should explicitly account for the human respiration, based on the spatial distribution of 12 13 the population.

14

One often stated objective of the top-down inversion of fossil fuel CO₂ emissions is to 15 provide an independent verification of the bottom-up estimates, i.e. the inventories (Levin et 16 al., 2011;McKain et al., 2012;Duren and Miller, 2012). However, information about the 17 18 spatial and temporal distribution of the emissions has to be used for inverse modelling to limit 19 aggregation errors on the overall budget. In our case, the number of monitoring stations is far 20 too small to independently invert the spatial distribution of the emissions. We have been able 21 to rely on the comprehensive distribution from AirParif. With a larger number of monitoring 22 stations, it may be possible to estimate some information about the flux spatial distribution, 23 but atmospheric transport is not a reversible process and some accurate information about the 24 spatial distribution will likely be needed, so that the atmospheric inversion cannot be seen as 25 independent from the inventories, but rather as a mean to verify or refine them. In addition, as long as the accuracy on the atmospheric transport makes does not allow using night-time or 26 27 morning measurements, it will not be possible to monitor the daily cycle of the emissions. Thus, the computation of daily or monthly fluxes requires some robust information about the 28 daily cycle that should rely on inventories. Thus, again, our top-down emission estimate is far 29 30 from independent from the bottom-up inventory.

Although the inversion procedure provides a posterior uncertainty estimate, one should 1 2 interpret this uncertainty with caution. Indeed, the mathematical framework used here relies on a number of hypotheses, some of which are crude approximations of the reality, such as 3 4 the spatial and temporal correlations in the flux uncertainties or the unbiased atmospheric 5 transport modelling. The impact of these assumptions has not been quantified. Although we have no "truth" to benchmark the inversion results, and there are not even enough 6 7 measurement sites to perform 'leave-one-out' tests, one can perform some sanity checks on 8 the results. One sanity check is the comparison of the measured and modelled mole fractions 9 (Figure 8 and Figure S4). The analysis of these figures confirms the ability of our inversion 10 to improve the measurement-model agreement. Nevertheless, we note that the posterior 11 misfit (≈ 2.5 ppm) is still a significant fraction of the signal that is analysed (10-20 ppm). The 12 crucial question is whether the atmospheric modelling error is random or a bias and we have 13 no element to answer that question. The other sanity check consists in analysing the validity of the retrieved daily fluxes (Figure 9). In this respect, the daily fluxes show day-to-day 14 15 variations that are suspicious, although not refutable at this stage. A result that points in favour of the flux inversions shown here is the significant reduction from the prior during a 16 period with temperatures above the seasonal normal, and the negative correction of the 17 18 emissions during November from the prior value that is based on an inventory simulating 19 January emissions. A single such event is certainly not sufficient to validate the inversion 20 We shall apply the same inversion setup to more than a year of system, however. 21 measurements and analyse the results with respect to the temperature anomaly or other shortterm event that may have a significant influence on the Île-de-France CO₂ emissions. More 22 23 measurement sites are needed to better evaluate the skill of the inversion. The deployment of 24 a network of 5 sites around Paris within the framework of the CarboCount-City project will help in this direction. In addition, inlet at different altitudes will be installed on the Eiffel 25 26 tower station for a better assessment of the CO₂ vertical distribution and transport within the urban area. These will be most useful for the longer-term objective of improving the 27 28 atmospheric transport modelling within the city, which may allow the EIF measurements to 29 be used by the inversion system.

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- 9

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1 Tables and captions

3	Table 1	: Information	about the CC	2 measuring	stations the	at are used in	this paper.
				- 0	/		1 1

Location	Acronym	Latitude [°]	Longitude [°]	Height AGL [m]	Distance from Paris centre [km]
Eiffel Tower	EIF	48.8582	2.2946	300	4 (W)
Montgé-en-Goële	MON	49.0284	2.7489	9	35 (NE)
Gonesse	GON	48.9908	2.4446	4	16 (N)
Gif sur Yvette	GIF	48.7100	2.1475	7	23 (SW)
Trainou Forest	TRN	47.9647	2.1125	180	101 (S)





Figure 1 : Map of the study area showing the location of the continuous CO₂ measurement
stations that are used in this paper (red dots). The black lines show the model grid with a 2
km resolution at the centre, and 10 km on the sides. The red line shows the limits of the Îlede-France region.

8



Figure 2 : Typical day-total CO_2 emissions of Île-de-France, according to AirParif year 2008 inventory, for a weekday in October. The point sources are not included in this map. The emissions are provided for the area outlined in red in Figure 1. The resolution is 1 km. The grid is 0.2° in latitude and 0.4° in longitude.



Figure 3: Temporal variation of the main CO₂ emission sectors according to the AirParif
inventory for the whole Ile de France region. The figure shows, for 5 typical months and 3
typical days (Weekday, Satuday, Sunday), the hourly CO₂ emissions. The black line is the
total emission (left scale) while the four coloured lines are for different sectors (right scale).







Figure 4: Mean diurnal cycle of the biogenic flux (Net Ecosystem exchange) for the 12
calendar months and for the same area as in Figures 2 and 3 which is outlined in red in Figure
1. The values were derived from an average of the C-Tessel simulations.



2 Figure 5: Time series of the measured (red) and modelled (green) CO₂ mole fraction [ppm] 3 for the five sites used in this paper (See Table 1). The black line is the modelled mole fraction 4 that is transported from the domain boundaries, with additional contribution from 5 anthropogenic emissions outside the Île-de-France region (Edgar fluxes). The green line 6 shows the modelled mole fraction that includes the same contributions, plus the biogenic 7 fluxes within the modelling domain and the anthropogenic emissions within the Île-de-France 8 Note that there are some time periods when no region. Red are the observations. 9 measurements are available due to either calibration processes or, more rarely, failure of the monitoring instrumentation. For such periods, modelling results are not shown. The symbols 10 11 show the mean of the afternoon measurement/model values that are used for the inversion. 12 The blue arrows indicate the wind speed and direction at noon. A length equivalent to 1 day 13 on the X-axis is for a wind speed of 10 m/s. Grey shaded areas indicate Sundays. This figure



390 0.73 R= 0.90 R= 0.84 R =0.61 R= 0.81 R =400 420 440 400 420 440 400 420 440 400 420 440 400 420 440 2010/11/27 2010/12/26 Tcor= 7 days 5 Obs. : GIF Obs. : GON Obs. : MON Obs. : EIF Obs. : TRN-180 6 Figure 6 : Scatter plot of the measured and modelled CO₂ mole fractions at the 5 monitoring

stations within and in the vicinity of the Paris city. The model vs measurement bias, standard
deviation and correlations are provided within each subplot. This figure is for the 30 days
period starting on 2010/11/27. Figure S2 in the supplementary shows the same figure for the
other period.



Figure 7 : Time series of the mole fraction differences between a station (Y-axis label) and another one used as a reference (either GIF or GON) and selected based on the wind direction (see section 3.3). The symbols show the mean afternoon concentrations (12AM-4PM) for the measurements (red), the prior (green) and the posterior (blue) estimates. As in Figure 5, the arrows indicate the wind speed and direction. A similar figure for the other time period is shown in the supplementary material.



2 Figure 8 : Scatter plot of the measured and modelled concentration gradients for 3 downwind 3 stations; either GIF or MON are used as an upwind reference The first row shows the mole 4 fraction simulated using the boundary conditions and the anthropogenic emissions outside Île-5 de-France (yF in equation 1) against the measurements. The second row shows the 6 concentration estimates derived from the prior values for the biogenic fluxes and 7 anthropogenic fluxes against the corrected measurements (i.e. y - yF in equation 1). The last 8 row is the same but using the posterior estimates. This figure is for the Nov-Dec period. A 9 similar figure for the other time period is shown in the supplementary material.

1



Figure 9 : Daily flux estimates of the anthropogenic emission for the 30 days of the period.
The blue line and shading shows the prior flux according to the AirParif inventory together
with its assumed uncertainty. Yellow shading indicate Sundays; note the weekly cycle with
lower values during Saturdays and Sundays. The red symbols and bars show the posterior
estimates with their uncertainty range. Both 30-day periods are shown.





Figure 10 : Total flux estimates over the full 30 day period, for the 4 6-hour periods. Red is
for the anthropogenic emissions, green is for the biogenic fluxes while blue is for the total.
The prior estimates are shown as open rectangles while the posterior are shown as filled
rectangles. Both 30-day periods are shown independently.

1 Supplementary material

2



4 Figure S- 1: Same as Figure 5 but for the 30 days period starting on October 21^{st} .



3 Figure S- 2 : Same as Figure 6 but for the 30 day period starting on October 21st.



Figure S- 3 : Measurement-Model difference in CO_2 mole fraction at the EIF site as a function of time, wind speed and direction. The position of the symbols indicates the wind direction (top-right is for a wind from the North-East) and speed (the circles indicate a wind of 10 m s⁻¹). The wind speeds have been bounded at 12 m s⁻¹.

10











7 Figure S-5 : Same as Figure 8 but for the 30-day period starting on October 21st.