

# CO<sub>2</sub> budgeting at the regional scale using a Lagrangian experimental strategy and meso-scale modeling

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**Abstract.** An atmospheric Lagrangian experiment for regional CO<sub>2</sub> budgeting with aircraft measurements took place during the CarboEurope Regional Experiment Strategy campaign (CERES) in south-west France, in June 2005. The atmospheric CO<sub>2</sub> aircraft measurements taken upstream and downstream of an active and homogeneous pine forest revealed a CO<sub>2</sub> depletion in the same air mass, using a Lagrangian strategy. This field experiment was analyzed with a meteorological meso-scale model interactively coupled with a surface scheme, with plant assimilation, ecosystem respiration, anthropogenic CO<sub>2</sub> emissions and sea fluxes. First, the model was carefully validated against observations made close to the surface and in the atmospheric boundary layer. Then, the carbon budget was evaluated using the numerous CERES observations, by upscaling the surface fluxes observations, and using the modeling results, in order to estimate the relative contribution of each physical process.

A good agreement is found between the two methods which use the same vegetation map: the estimation of the regional CO<sub>2</sub> surface flux by the Eulerian meso-scale model budget is close to the budget deduced from the upscaling of the observed surface fluxes, and found a budget between  $-9.4$  and  $-12.1 \mu\text{mol.m}^{-2}.\text{s}^{-1}$ , depending on the size of the considered area. Nevertheless, the associated uncertainties are rather large for the upscaling method and reach 50%. A third method, using Lagrangian observations of CO<sub>2</sub>

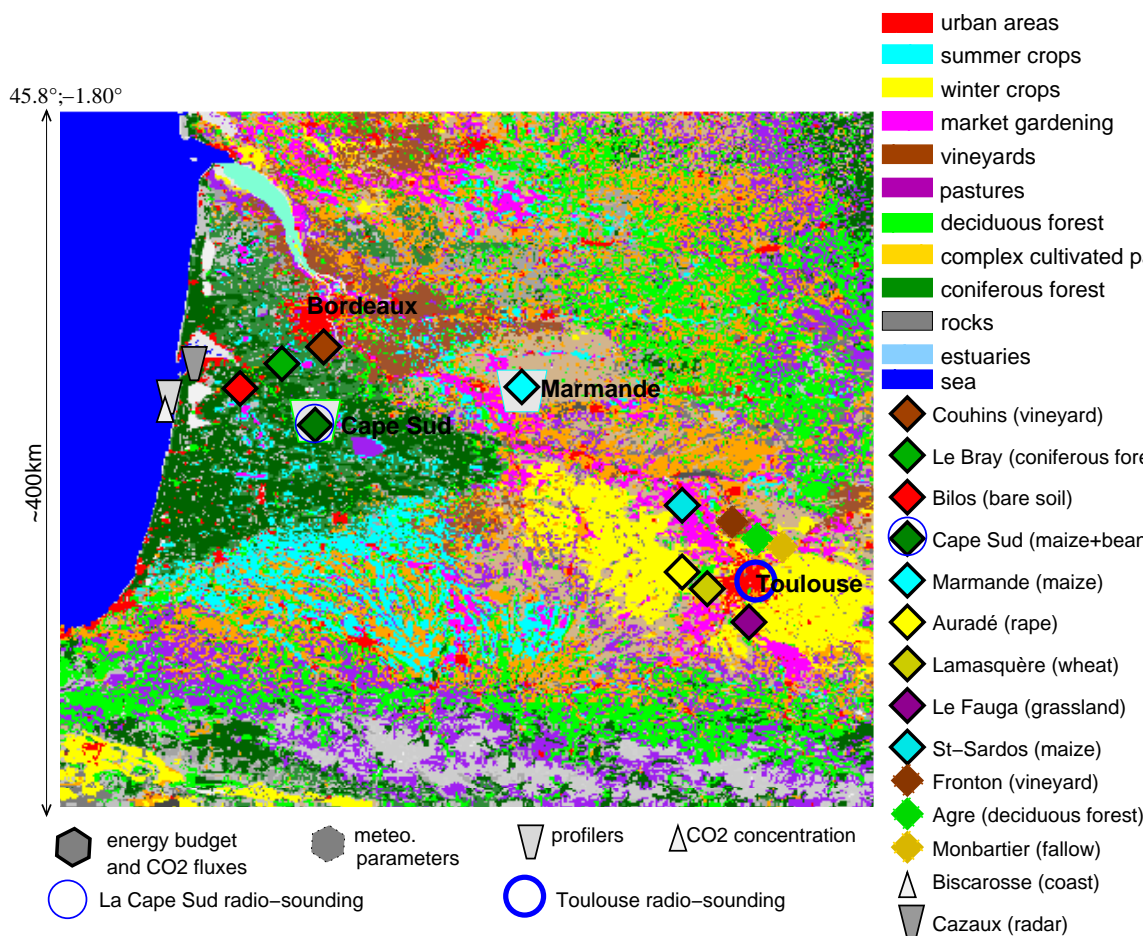
estimates a regional CO<sub>2</sub> budget a few different and more scattered, ( $-16.8 \mu\text{mol.m}^{-2}.\text{s}^{-1}$  for the small sub-domain and  $-8.6 \mu\text{mol.m}^{-2}.\text{s}^{-1}$  for the larger one). For this budgeting method, we estimate a mean of 31% error, mainly arising from the time of integration between the two measurements of the Lagrangian experiment. The paper describes in details the three methods to assess the regional CO<sub>2</sub> budget and the associated errors.

## 1 Introduction

As the major greenhouse gas actor in climate change, a better knowledge of regional atmospheric CO<sub>2</sub> budget is needed, as well as an understanding of underlying processes. At the global and continental scale, models are able to infer the CO<sub>2</sub> surface fluxes (Bousquet et al., 1998), but the resolution of global inversions is too coarse to get accurate information at the regional scale. Meteorological meso-scale models permit the use of higher spatial resolution, better Atmospheric Boundary Layer (ABL) and surface energy budget parametrizations, and the explicit resolution of local to regional wind circulations that are crucial for the understanding of the atmospheric CO<sub>2</sub> variability (Pérez-Landa et al., 2007). Previous studies (Sarrat et al., 2007a,b; Nicholls et al., 2004; Denning et al., 2003) showed the capacity of meso-scale models to simulate the CO<sub>2</sub> surface fluxes as well as the atmospheric concentration gradients and their variability. In this paper, we use regional modeling and experimental data from the CarboEurope Regional Experiment Strategy



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**Fig. 1.** Experimental set-up during the CERES 2005 campaign, over the land cover classification.

(CERES) campaign (Dolman et al., 2006) to estimate the CO<sub>2</sub> uptake fluxes during daytime, over a flat and homogeneous active forest, surrounded by an agricultural area.

A specific Lagrangian experiment was conducted in June 2005, during a westerly wind episode, over the Landes forest, south-western France (Fig. 1). The Lagrangian strategy consisted of following an oceanic air mass moving towards land and sampling it, first above the ocean, then over the active forest and then again further downstream of the forest, knowing the wind speed and the time needed for the air mass to be advected. The measured CO<sub>2</sub> decrease between upstream and downstream of the forest is related to the CO<sub>2</sub> uptake.

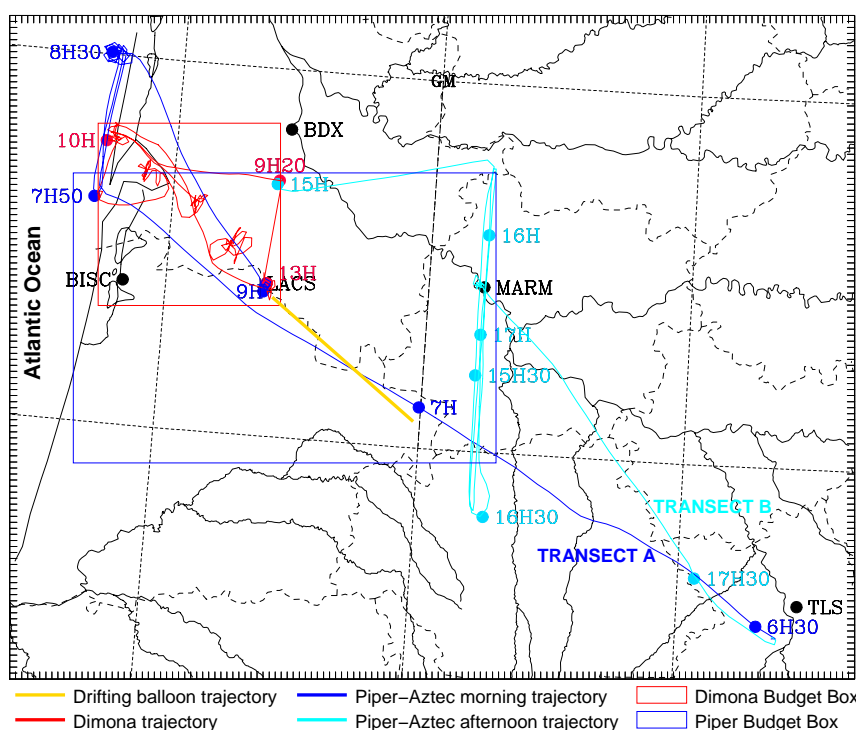
Lagrangian aircraft experiments have recently been established as a method to measure regional scale trace gas fluxes (Lin et al., 2004, 2006, 2007; Matross et al., 2006). Schmitgen et al. (2004) already showed the potential of a Lagrangian budgeting approach for accurately estimating CO<sub>2</sub> fluxes for the Landes region, using the aircraft Lagrangian observations and a simple Lagrangian model.

Here, in this study, both experimental data from the surface and the aircraft observations and a meteorological meso-scale model at high resolution are used. After a description of the experimental and modeling tools, the simulation outputs are compared to several types of observations. Then, we analyze the CO<sub>2</sub> regional budget using both the modeling results and the Lagrangian experiment, in order to quantify the regional CO<sub>2</sub> fluxes.

## 2 Method to estimate CO<sub>2</sub> budget at the regional scale

### 2.1 The Lagrangian Experiment Strategy

The May–June 2005 CERES experiment was performed in south-western of France, as shown in Fig. 1. The area contains a large and rather homogeneous pine forest (the Landes forest) bounded by the Atlantic ocean to the west and an agricultural area to the east. This area was chosen because the previous HAPEX-MOBILHY experiment (André et al., 1986) dedicated to evapotranspiration and land



**Fig. 2.** Trajectories of the aircraft and balloon: red is the Dimona trajectory, dark and light blue are the two Piper-Aztec trajectories, respectively in the morning (Transect. A) and in the afternoon (Transect. B). The red and blue sub-domains are the Dimona and Piper budget boxes. The yellow line corresponds to the drifting balloon trajectory, giving the direction and wind speed during the experiment.

surface-atmosphere interactions offered a large database and many useful studies (e.g. Bélair et al., 1998; Habets et al., 1999).

During CERES 2005, the experimental set-up included 10 surface flux stations, located in ecosystems representative of the region (vineyards, maize, wheat, pine forest...). In the pine forest, the evolution of the Atmospheric Boundary Layer (ABL) was monitored at La Cape Sud (denoted LACS hereafter) with radio-soundings, a UHF radar, meteorological and flux measurements. The vertical and horizontal variations of CO<sub>2</sub> concentration were sampled by four research aircraft and two towers (in Biscarosse and Marmande, respectively noted BISC and MARM hereafter). More details of the experimental strategy can be found in Dolman et al. (2006).

Two Lagrangian experiments were performed jointly on the 6 of June 2005, by Piper-Aztec and Dimona aircraft. The north-west wind was regular all day, with a speed of nearly  $6.5 \text{ m.s}^{-1}$  in the ABL, and was spatially homogeneous. A few clouds were observed near the Atlantic coast, but elsewhere the sky was clear and the temperature reached 25°C.

The Piper-Aztec sampled the air mass in the early morning, close to the Atlantic coast (Transect. A, as shown in Fig. 2). The same air mass was sampled once again (Transect. B), a few hours later, in the afternoon, over land by the same aircraft.

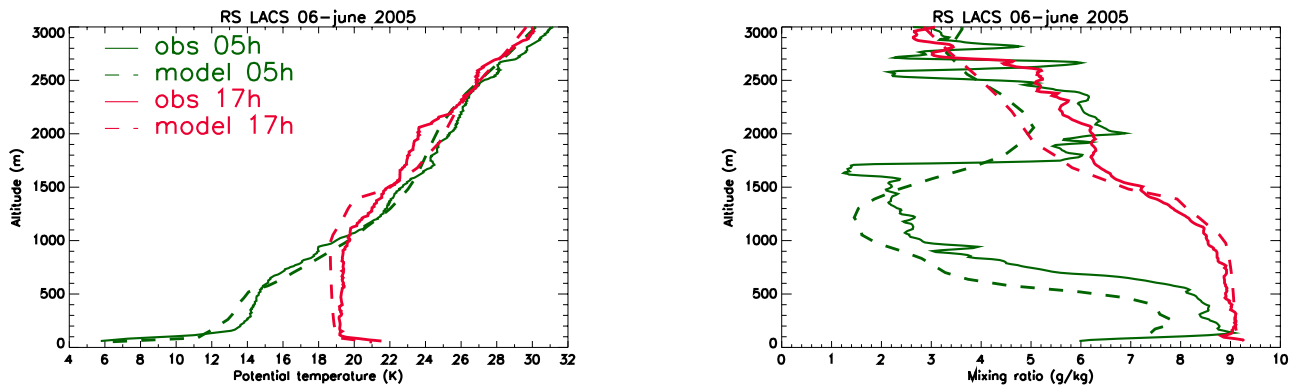
By this time, the same strategy was applied by the Dimona, but over a shorter window. This aircraft sampled the air mass over the Atlantic Ocean at 10:00 UTC, followed this air mass during three hours over land and sampled the CO<sub>2</sub> concentration over vertical profile at LACS (red trajectory in Fig. 2).

The two aircraft did not cover the same area: the Piper-Aztec flew for longer and over a larger domain, encompassing the whole coniferous forest and cropland, while the area flown by the Dimona was more homogeneous and essentially dominated by forest.

Two special constant volume balloons were launched from LACS to sample the temperature and moisture in a truly Lagrangian fashion at fixed density levels. Information on the wind patterns and flight paths of the balloons were transmitted to the pilots of the aircraft in order to help them fly in a Lagrangian sampling mode.

## 2.2 Meso-scale modeling with Meso-NH

This Lagrangian experiment day was simulated using the meteorological model, Meso-NH, a non-hydrostatic meso-scale model (Bélair et al., 1998). The model was run with two domains (two-way nesting mode) allowing a high resolution of 2 km for the inner domain, and a 10 km resolution for the larger one (domain width 900 km × 900 km). The atmospheric Meso-NH model includes the CO<sub>2</sub> concentration, transported as a passive scalar.



**Fig. 3.** Potential temperature (°C) (left) and mixing ratio (right) observed and simulated in the forest central site La Cape Sud at 05:00 UTC (green) and 17:00 UTC (red).

The surface energy and CO<sub>2</sub> fluxes are computed on-line by the surface scheme, ISBA-A-gs (Noilhan and Planton, 1989; Calvet et al., 1998), including CO<sub>2</sub> assimilation by the vegetation and ecosystem respiration. In the surface scheme, the latent heat flux as well as the carbon flux are computed through the stomatal conductance variable.

The land cover is from an improvement of the Ecoclimap database at 1 km resolution (Champeaux et al., 2005; Masson et al., 2003) and contains 62 types of cover. For the natural surface (i.e. all surface types but town, sea and lake), a tile approach is used in ISBA-A-gs, in which each 2 km grid cell is divided into patches (bare soil, snow, rock, tree, coniferous, evergreen, C3 crops, C4 crops, irrigated crops, grassland tropical grasslands and parks). The energy and CO<sub>2</sub> budgets are calculated for each patch within each grid cell and then averaged at the grid scale, according to the areal fraction of each patch.

Some important parameters needed to compute the surface processes are tabulated in the surface model, for these 12 vegetation types. The parameters are the surface roughness length ( $z_0$ ), the root zone depth ( $d_2$ ), the mesophyll conductance ( $g_{mes}$ ), and the ecosystem respiration at 25°C ( $RE_{25}$ ). All of these parameters have been calibrated independently in the framework of previous studies. The values of  $z_0$  and  $d_2$  were calibrated from detailed validation over France in the hydro-meteorological model SIM (see Habets et al., 2008 for a detailed description). The main parameters concerning the carbon cycle in the model are the  $g_{mes}$  and  $RE_{25}$  parameters. A specific calibration was done for these parameters, using data from previous campaigns dedicated to atmospheric regional CO<sub>2</sub>, as the COCA Experiment in the Landes forest, 2001 (Schmitgen et al., 2004) and RE-CAB in Netherlands and Spain in 2003 (Pérez-Landa et al., 2007). These datasets offered the opportunity to calibrate these two parameters against CO<sub>2</sub> surface fluxes and atmospheric concentration. All these calibrations are independent of this study and have been kept for this simulation. They

were done operating a 1-D version of the meso-scale model, with on-line coupling of the CO<sub>2</sub> between the surface and the atmosphere.

The Leaf Area Index (LAI) was taken from the remote sensing observations of the Normalized Difference Vegetation Index (NDVI) from the MODIS sensor (Roujean and Lacaze, 2002).

The anthropogenic CO<sub>2</sub> emissions were provided by the 10 km inventory of the Stuttgart University (Dolman et al., 2006). Oceanic CO<sub>2</sub> fluxes were parametrized according to Takahashi et al. (1997).

The simulation was initialized at 18:00 UTC on the 5 of June, using the European Centre for Medium-Range Weather Forecasts (ECMWF) analysis, for both the surface, and the meteorological fields and run for 30 h. The meteorological lateral boundary conditions are forced every six hours with the ECMWF analysis. The CO<sub>2</sub> concentration were initialized with a homogeneous vertical profile over the whole domain, while a zero horizontal concentration gradient was applied at the boundaries of the larger scale domain.

### 2.3 Three methods for estimating the CO<sub>2</sub> budget

In order to establish the regional CO<sub>2</sub> budget, as accurately as possible, we used both the meso-scale modeling results and the set of observations made during the Lagrangian experiment.

The estimation of the CO<sub>2</sub> budget is based on the resolution of the equation of the CO<sub>2</sub> temporal evolution decomposed into three terms:

$$\frac{\partial \text{CO}_2}{\partial t} = -\bar{u} \frac{\partial \text{CO}_2}{\partial x} - \bar{v} \frac{\partial \text{CO}_2}{\partial y} - \frac{\partial \overline{w' \text{CO}_2'}}{\partial z} \quad (1)$$

Where  $\overline{w' \text{CO}_2'}$  is the vertical turbulent flux of CO<sub>2</sub>,  $\bar{u}$  and  $\bar{v}$  the mean horizontal wind components.



We tested three methods to determine the regional CO<sub>2</sub> budget, based on the two aircraft measurements (Dimona and Piper-Aztec described before):

1. After validation, the Meso-NH model was used to estimate the contribution of each transport term, giving a numerical Eulerian budget (Sect. 3.2);
2. The observations from the flux sites network were used to calculate an average regional “bottom-up” flux using a simple averaging based upon the fractional coverage of each ecosystem (Sect. 3.3);
3. The Lagrangian budget was estimated using the aircraft observations (Sect. 3.4).

For each of the two aircraft experiments described in Sect. 3.1.4, the Dimona and the Piper-Aztec flights, we determined a sub-domain corresponding to the aircraft trajectories. In these sub-domains, the CO<sub>2</sub> surface fluxes were evaluated using the 3 methods.

### 3 Results

#### 3.1 Comparisons of model and observations

In order to validate the simulation, the Meso-NH outputs were compared with several types of observations made during CERES, including the ABL height and structure, the CO<sub>2</sub> surface fluxes for 6 different cover types, the CO<sub>2</sub> concentration near the coast and in the agricultural zones. Finally, the observations of CO<sub>2</sub> concentration made during the Lagrangian flights within the ABL were compared with the model outputs.

##### 3.1.1 Atmospheric Boundary Layer

During the CERES Intensive Observation Period (IOP), radio-soundings (RS) were launched every 3 h at the central forest site LACS. Figure 3 shows an evaluation of the simulated ABL height compared with the RS launched during the early morning, at 05:00 UTC and at 17:00 UTC. This figure compares the vertical profiles of the potential temperature (left) and of the mixing ratio (right). At 05:00 UTC, the stable ABL is well simulated by the model, even if the mixing ratio is underestimated: the vertical structure within the first 2000 m is in good agreement with the observations.

At 17:00 UTC, the potential temperature is slightly underestimated at the top of the ABL, but the vertical profile of the mixing ratio shows that the simulated ABL height is at almost 1100 m, in agreement with the observations. During the day, the humidity in the ABL increases and reaches 9 g.kg<sup>-1</sup>, due to the evapotranspiration and the moisture advection from the sea.

##### 3.1.2 CO<sub>2</sub>, latent and sensible heat surface fluxes

The diurnal cycles of the measured CO<sub>2</sub> fluxes are compared for the 6 of June, with the simulated values in Fig. 4 at several surface stations with various vegetation cover types: rapeseed, wheat, pine forest, vineyard and maize. The flux sites were selected to be representative of the main land cover types of the area.

In general, the model is in good agreement with the CO<sub>2</sub> observations, the nocturnal respiration and the diurnal NEE (Net Ecosystem Exchange).

At this time of the year, the winter crops (rapeseed and wheat) assimilate carbon at close to their maximal rates (up to 25 μmol.m<sup>-2</sup>.s<sup>-1</sup>), as does the pine forest. Although the assimilation differs between two maize site, the model is able to simulate both of them correctly, mainly because the LAI field prescribed in the meso-scale model has a good level of realism.

When comparing the model with observations of latent and sensible heat fluxes, a weaker agreement is found than for CO<sub>2</sub> fluxes, as shown in Fig. 5. In the western part of the domain, due to the cloud cover, it is more difficult to compare the model results at the pine forest and the vineyards sites. However, for these both sites, the sensible heat flux is reasonably well simulated. A similar behaviour is noted at the eastern sites (wheat and maize) less affected by the cloudiness: the latent heat flux is overestimated by the model while the sensible heat flux is better.

At this stage, it is difficult to make conclusions about this model failure, since it is well known that the Eddy Covariance technique used for these measurements tends to underestimate the surface turbulent fluxes.

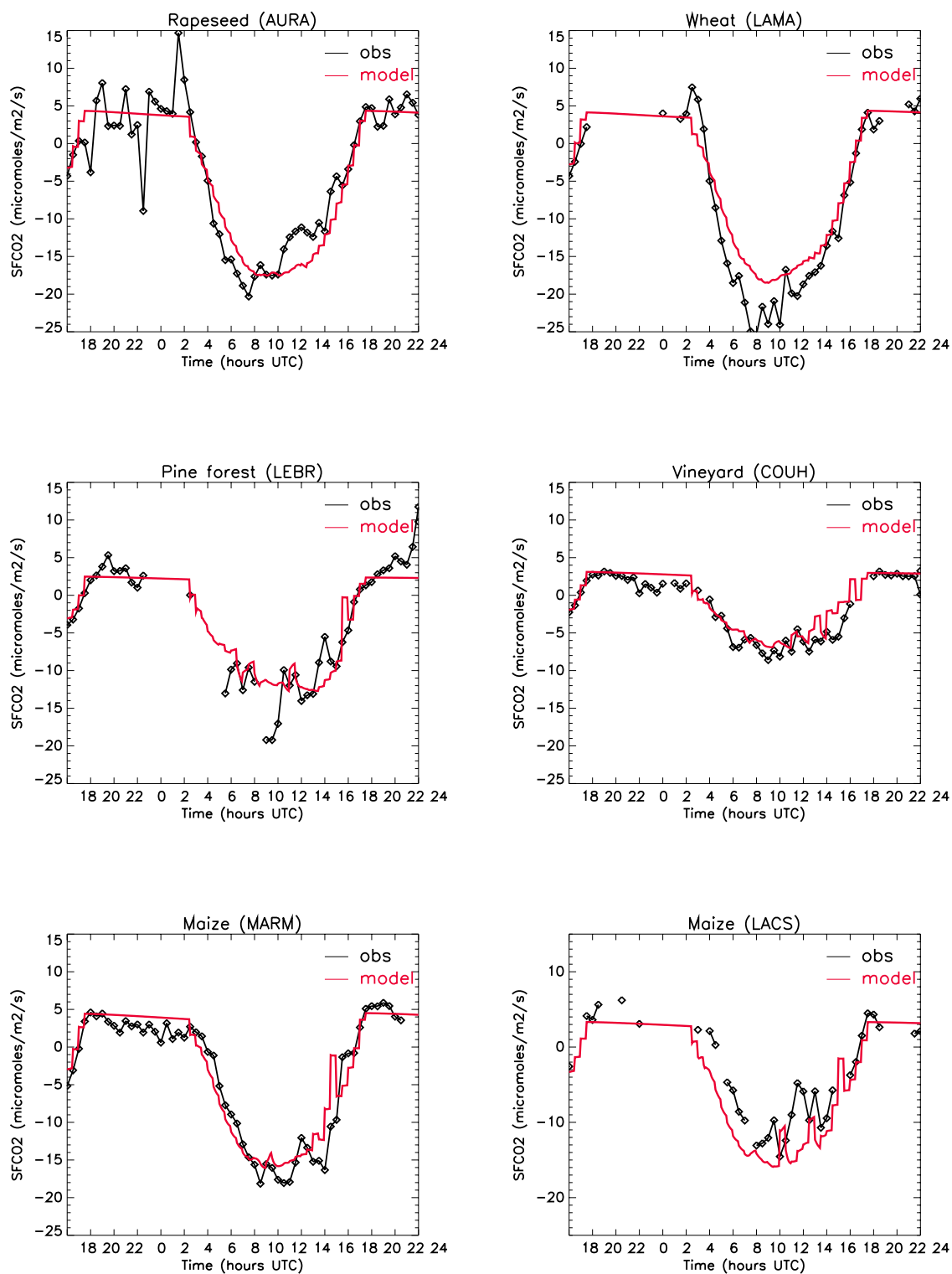
##### 3.1.3 CO<sub>2</sub> concentration at the tower sites

The observed CO<sub>2</sub> concentration at the BISC and MARM sites are compared to the simulated concentration in Fig. 6.

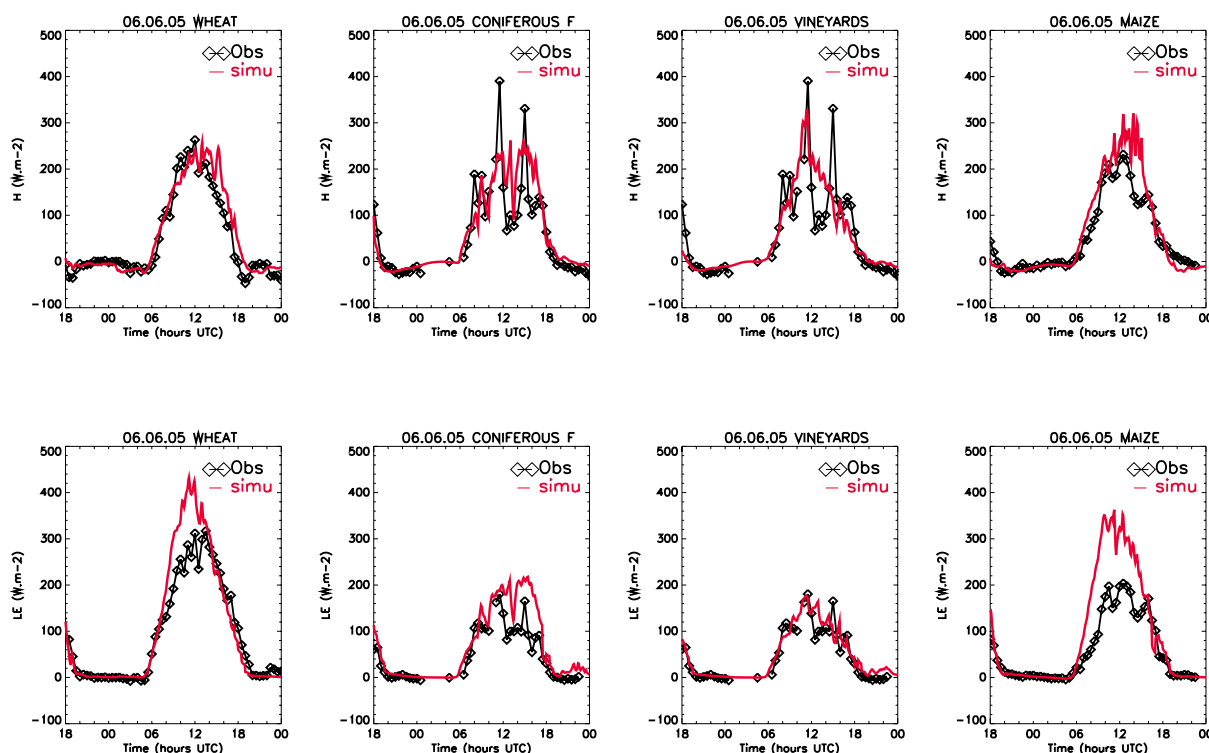
Although the CO<sub>2</sub> concentration is slightly overestimated at night at BISC (left panel), the simulated concentration after 06:00 UTC is in good agreement with the observations. These comparisons show that the CO<sub>2</sub> inflow is steady state and regular all day.

At MARM (right panel), the nocturnal maximum due to the ecosystem respiration is underestimated by the model. This discrepancy may occur because the first level of the model is at 20 m height, while the surface measurements are representative of the ground-level concentration. The daily concentration is better represented by the model, when diurnal mixing occurs.

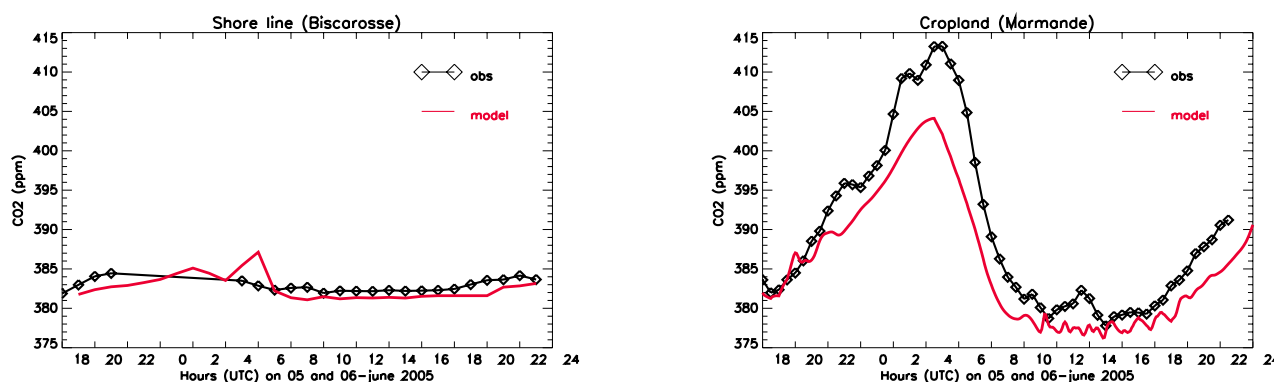
The difference in concentration for both the observed and the modeled and between the ocean and the agricultural areas during the afternoon is approximately 4 to 6 ppm.



**Fig. 4.** Comparisons of CO<sub>2</sub> surface fluxes ( $\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) at several surface stations: observations (black), model (red). The comparisons begin on June 2005 at 18:00 UTC till the June 2006 all day long.



**Fig. 5.** Comparisons of sensible heat flux (top) and latent heat fluxes (bottom) ( $\text{W.m}^{-2}$ ) at the wheat (Lamasquère), coniferous forest (Le Bray), vineyards (Cuhins) and maize (Marmande) stations; observations (black), model (red). The comparisons begin on June 2005 at 18:00 UTC till the June 2006 all day long.



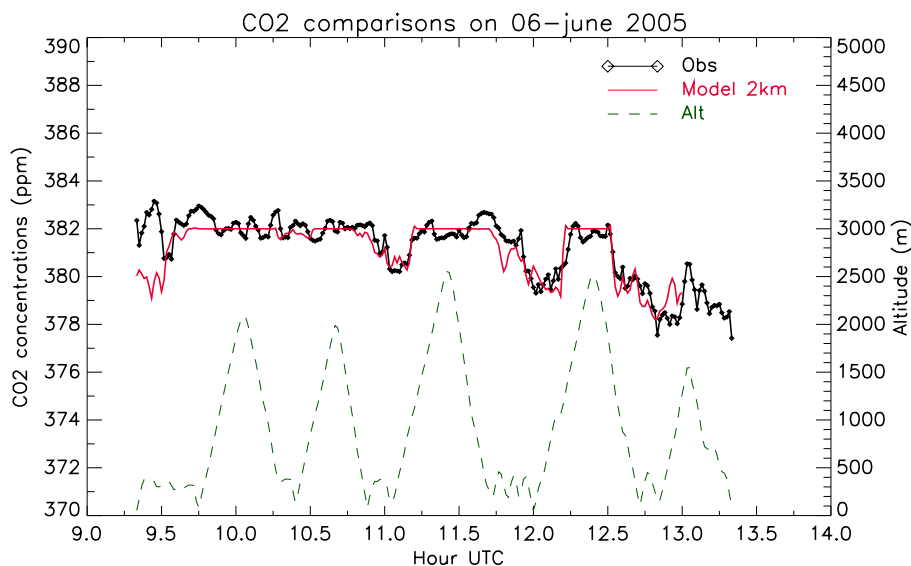
**Fig. 6.** CO<sub>2</sub> concentration simulated and observed in BISC near the Atlantic shore line (left) and in MARM in the agricultural area (right).

### 3.1.4 Aircraft in-situ measurements

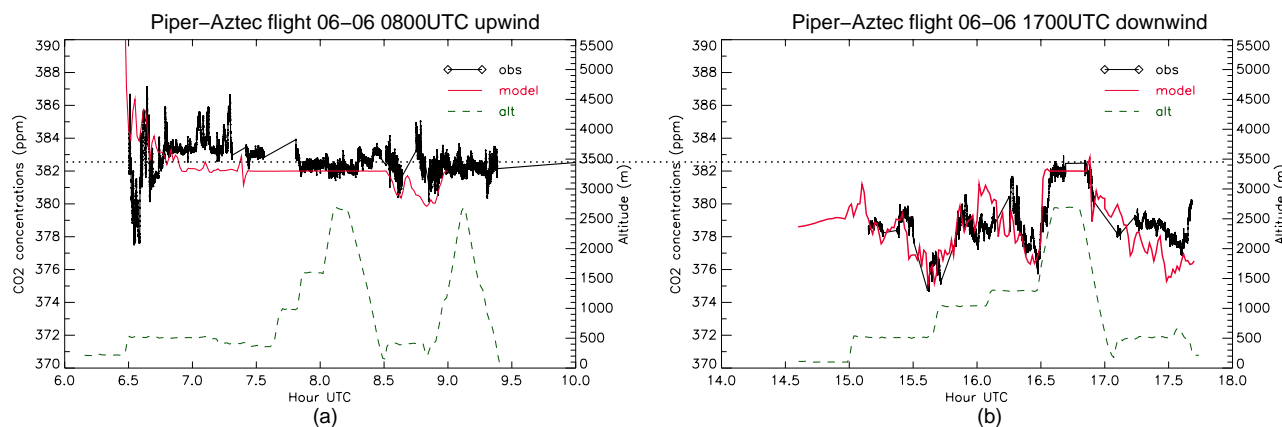
During this IOP day, several measurement flights were operated, over the Landes forest.

The MetAir Dimona aircraft (<http://www.metair.ch>) measured CO<sub>2</sub> at 1 Hz with an accuracy of better than 0.5 ppm using a combination of open and closed path sensors, as well as flask analysis (Dolman et al., 2006), along a trajectory following the air mass for over 3.25 h which is displayed in Fig. 2. The aircraft trajectory is simulated on-line in the

model, which means that the CO<sub>2</sub> in the model is interpolated in space and time to the exact location of the aircraft trajectory. The comparison between the CO<sub>2</sub> observed by the Dimona and the simulation (Fig. 7) shows that the model is able to reproduce slight concentration variabilities in and above the ABL, in the morning and the early afternoon, due to the vertical mixing and the CO<sub>2</sub> assimilation by the vegetation. The model also reproduces the slow decrease of CO<sub>2</sub> concentration from the morning to the time at the end of the flight.



**Fig. 7.** CO<sub>2</sub> concentration (ppm) time series: from the model (red) and aircraft observations (black), altitude (green dashed lines) during the Dimona flight over the Landes region. The model outputs are extracted on-line during the run, at the exactly the same altitude, longitude, latitude and time aircraft position.



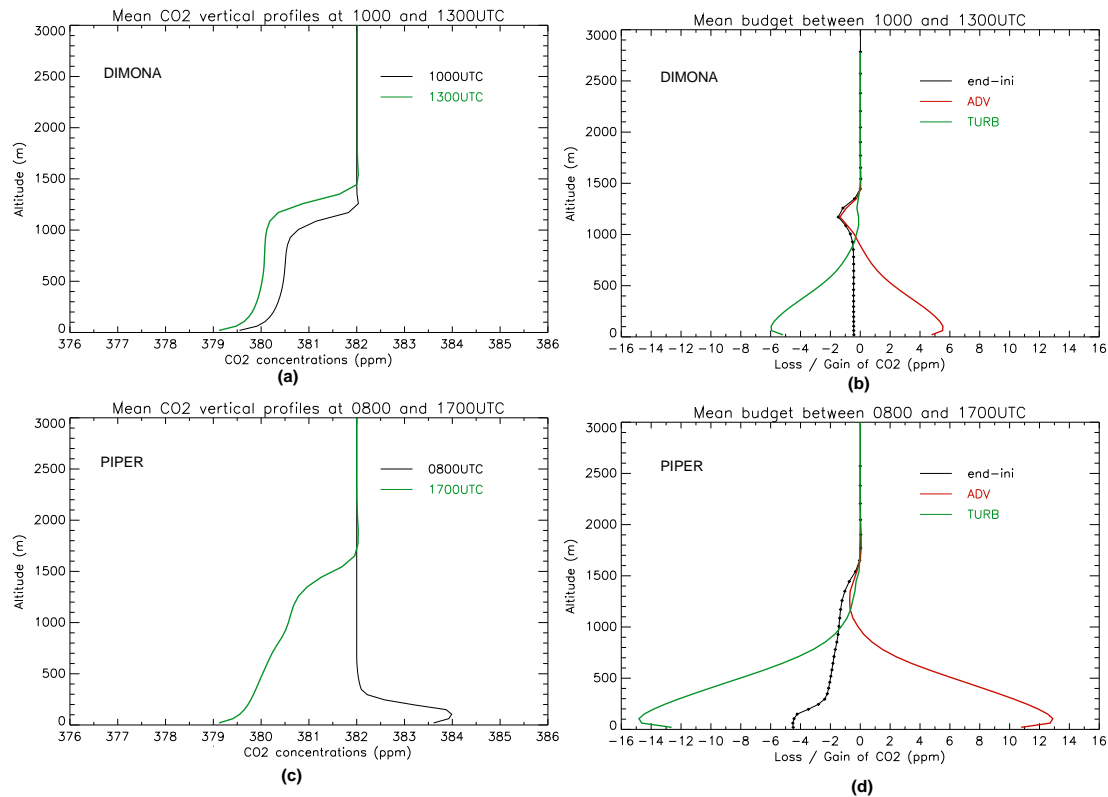
**Fig. 8.** CO<sub>2</sub> concentration time series: from the model (red) and aircraft observations (black), altitude (dashed lines). **(a)** for the upstream morning flight, **(b)** for the downstream afternoon flight. The model outputs are extracted on-line during the run, at the exactly the same altitude, longitude, latitude and time aircraft position.

A second aircraft, the Météo France Piper-Aztec, flew over the ocean in the morning (dark blue trajectory in Fig. 2) and then downstream to the Landes forest in the afternoon (light blue trajectory in Fig. 2), for the so-called “Lagrangian Experiment”, described in Sect. 2.1. The Fig. 8 shows the variation of CO<sub>2</sub> concentration between the transects A and B, observed by the aircraft measurements. The CO<sub>2</sub> time series show that during the morning flight, over the ocean and the forest area, the CO<sub>2</sub> concentration were constant with time and space: between 382 and 383 ppm in and above the ABL (Fig. 8a). During the afternoon flight, the CO<sub>2</sub> concentration was lower in the ABL, and remained close to 382 ppm above

it (Fig. 8b). Therefore, in the same air mass, the CO<sub>2</sub> depletion in the ABL upstream and downstream the pine forest is measured to be 4 to 6 ppm. The CO<sub>2</sub> concentration simulated by Meso-NH are in good agreement with these observations, although slight discrepancies occur at low altitudes.

The model outputs also compare reasonably well with other types of observations (air temperature, wind speed, surface energy fluxes), but these comparisons are not shown here. Moreover, previous studies (Sarrat et al., 2007a,b) showed that the model is able to reproduce the energy surface fluxes as well as the meso-scale circulations on the 27 May and 6 June, during CERES.





**Fig. 9.** Eulerian budget from the model: (a) averaged vertical profiles of CO<sub>2</sub> concentration over the Dimona sub-domain at 10:00 (black) and 13:00 UTC (green); (c) same than (a) for the Piper-Aztec sub-domain at 08:00 and 17:00 UTC; (b) and (d) averaged vertical profiles of the budget terms: turbulent tendency in green, advective tendency in red and difference between the morning and afternoon profiles in black, respectively for the Dimona and Piper-Aztec boxes.

### 3.2 Numerical Eulerian budget from Meso-NH

The Meso-NH model calculates the contribution of each term corresponding to the right-hand side of the Eq. (9).

In order to facilitate the analysis, the simulated budget terms are designated in the following as:

- the total advective tendency:  $ADV = -\bar{v} \frac{\partial CO_2}{\partial y}$ ,
- the turbulent tendency:  $TURB = -\frac{\partial \overline{w'CO_2'}}{\partial z}$ .

Therefore we have :  $\frac{\partial CO_2}{\partial t} = ADV + TURB$

The numerical budget terms are calculated separately for the two Lagrangian experiments, of the Piper-Aztec and the Dimona flights over their respective sub-domains. For the Dimona, the budget terms are integrated over the 3.25 h of the flight, between 10:00 and 13:00 UTC, over a sub-domain encompassing the aircraft trajectory (hereafter called the Dimona box, represented in Fig. 2).

Figure 9a shows the simulated CO<sub>2</sub> vertical profiles at 10:00 (black) and 13:00 UTC (green) averaged over the Dimona box, while Fig. 9b shows the horizontally averaged

budget terms. Near the surface, the turbulent flux due to assimilation is compensated for by the advection of CO<sub>2</sub>-rich oceanic air. At the top of the boundary layer, the turbulent flux is negligible.

Integrating the turbulent tendency  $TURB = -\frac{\partial \overline{w'CO_2'}}{\partial z}$ , from the surface up to top of the ABL gives the mean surface flux calculated by the model,  $\overline{w'CO_2'}_{BUDGET} = -11.5 \mu\text{mol.m}^{-2}.\text{s}^{-1}$ .

The equivalent integration is done for the Piper-Aztec flights, between 08:00 and 17:00 UTC and then averaged over the horizontal sub-domain (hereafter called the Piper box, displayed in Fig. 2).

Figure 9c shows the averaged vertical profiles of CO<sub>2</sub> concentration over the Piper box at 08:00 (black) and 17:00 UTC (green). The morning profile shows an accumulation of CO<sub>2</sub> near the surface due to the ecosystem respiration within the nocturnal stable ABL. At 17:00 UTC, the CO<sub>2</sub> depletion is very large near the surface (6 ppm) and varies between 2 and 6 ppm in the mixed ABL. This is in good agreement with the depletion observed by the Piper-Aztec aircraft during the Lagrangian flights (Fig. 8).

**Table 1.** Weighting of the CO<sub>2</sub> surface fluxes between 08:00 and 17:00 UTC and between 10:00 and 13:00 UTC ( $\mu\text{mol.m}^{-2}.\text{s}^{-1}$ ) observed at several CERES stations. The vegetation type percentage in the 2 boxes are deduced from the Ecoclimap database. The bare soil corresponds to 4 to 5% of each box, the remaining percentage of the surface corresponds to the fraction of sea, rivers, lakes or urban areas.

	Type of vegetation					Weighted total flux $\sum_i^{\text{obs}} C_i \overline{w'CO'_{2i}}$ $\mu\text{mol.m}^{-2}.\text{s}^{-1}$
	Tree	Coniferous	C3 crops	C4 crops	Grass	
Representative station	Couhins	Le Bray	Cape Sud beans Auradé Lamasquère	Cape Sud maize Marmande	Le Fauga	
Mean CO <sub>2</sub> flux 10:00–13:00 UTC	−7.4	−14.3	−11.2	−11.8	−4.3	−12.1
Mean CO <sub>2</sub> flux 08:00–17:00 UTC	−6.5	−11.8	−10.2	−10.9	−3.0	−9.4
Percent of the DIMONA box	10	68	4	5	3	
Percent of the PIPER box	18	51	7	6	7	

Figure 9d shows the vertical profiles of the advective (red) and turbulent (green) CO<sub>2</sub> tendencies, averaged between 08:00 and 17:00 UTC over the Piper box. The advective term tends to provide oceanic air which is higher in CO<sub>2</sub> than the ABL exposed to forest uptake. This enrichment is compensated for by the turbulent tendency (corresponding to the assimilation by the vegetation and the vertical mixing) which decreases the CO<sub>2</sub> in the layer especially near the surface.

At the top of the ABL, the simulated entrainment flux is once again negligible.

If we examine the budget term at the first level of the model, the net difference between 08:00 and 17:00 UTC is −6 ppm due to:

- the advection up to 13 ppm from oceanic CO<sub>2</sub>-rich air.
- a turbulent contribution, dominated by plant assimilation and resulting in a CO<sub>2</sub> variation that reaches −15 ppm near the surface.
- The integration over the ABL of the turbulent flux gives  $\overline{w'CO'_{2\text{BUDGET}}} = -10.4 \mu\text{mol.m}^{-2}.\text{s}^{-1}$ .

The anthropogenic emissions represent less than 2% of the total CO<sub>2</sub> fluxes at the surface, and thus they are negligible when averaged over the Piper or Dimona boxes.

### 3.3 CO<sub>2</sub> area-average surface fluxes from in-situ measurements

A second way to estimate the CO<sub>2</sub> surface fluxes over the region is to use the continuous flux observations at eddy covariance stations distributed in the area (Fig. 1). The surface station network is assumed to be dense enough during CERES to have a representative sample of the land cover and to upscale the net ecosystem exchange of the domain.

Table 1 lists the different flux stations and the fraction of the corresponding type of cover in the respective sub-domains. These proportions are deduced from the Ecoclimap land cover database.

The weighted observed fluxes are integrated between 10:00 and 13:00 UTC for the Dimona sub-domain, which is dominated by pine forest (68% of the box).

In the Dimona box, the weighted surface flux is  $\sum_i^{\text{obs}} \overline{w'CO'_{2i}} = -12.1 \mu\text{mol.m}^{-2}.\text{s}^{-1}$ . This value is in good agreement with the model Eulerian budget.

For the Piper-Aztec sub-domain, the mean fluxes are calculated between 08:00 and 17:00 UTC. In this box, the weighted surface flux is  $\sum_i^{\text{obs}} \overline{w'CO'_{2i}} = -9.4 \mu\text{mol.m}^{-2}.\text{s}^{-1}$ , a value slightly lower than the model Eulerian budget.

The weighted CO<sub>2</sub> flux over the Dimona sub-domain is logically higher than the flux over the Piper-Aztec sub-domain, because the hours of integration around noon are the most favourable for the photosynthetic activity. Moreover, the Dimona box is dominated by the pine forest which is relatively active at this time of the year.

### 3.4 Lagrangian budgeting

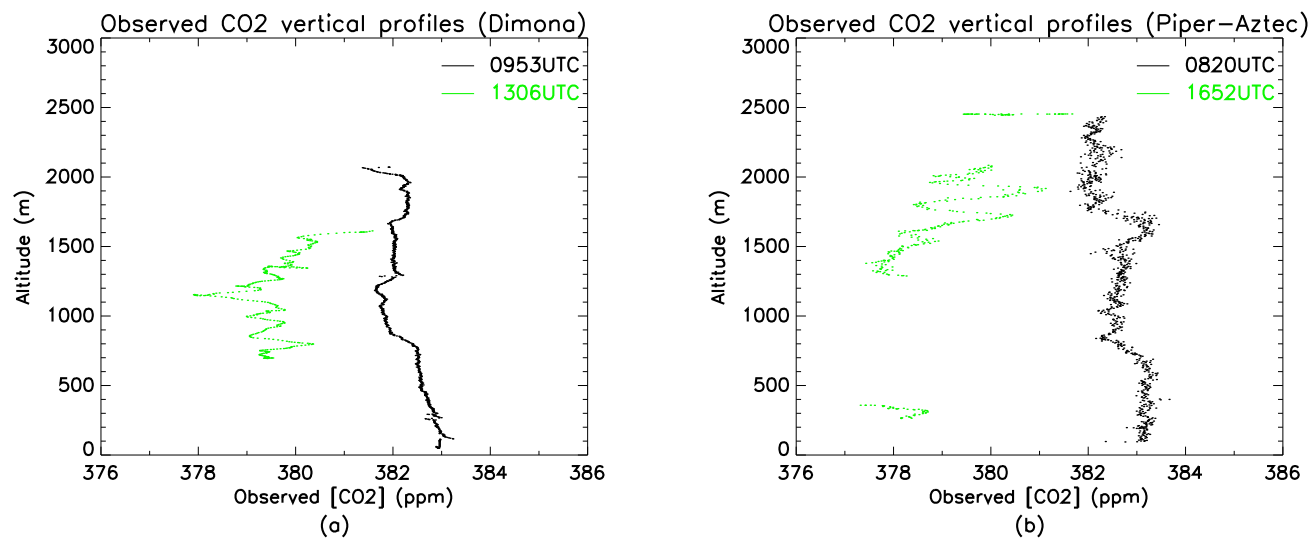
The third way to establish a regional budget is to use the aircraft observations. Integrating Eq. (1) from the surface to above the ABL height ( $z_{\text{max}}$ ) leads to:

$$\int_0^{z_{\text{max}}} \frac{\partial \text{CO}_2}{\partial t} dz = -[\overline{w'CO'_{2z_{\text{max}}}} - \overline{w'CO'_{2\text{surf}}}] \quad (2)$$

Where  $\overline{w'CO'_{2z_{\text{max}}}} = 0$ , before the turbulent fluxes are null above the ABL (here  $z_{\text{max}} > z_i$  the ABL height). The entrainment flux is not expressed here since the integration is done between the surface up to  $z_{\text{max}}$  which is above the ABL height.

As we consider the same air mass during its displacement, the advection is neglected. We can deduce from Eq. (2), the CO<sub>2</sub> flux at the surface,  $\overline{w'CO'_{2\text{surf}}}$ .

By this way, we obtain the following term:  $\text{LAG}_{\text{obs}} = \int_0^{z_i} \frac{\partial \text{CO}_2}{\partial t} dz$ . It corresponds to the variation of CO<sub>2</sub> into the same air mass observed during the morning flights near the coast line and the observations made in the afternoon further inland, for the two aircraft.



**Fig. 10.** Vertical profiles of CO<sub>2</sub> concentration observed by: the Dimona aircraft at 09:53 over the ocean (black) and 13:06 UTC inland (green) (a); and the Piper-Aztec aircraft at 08:20 UTC over the ocean (black) and 16:52 UTC inland (green) (b).

**Table 2.** CO<sub>2</sub> budget calculated according 3 methods (in  $\mu\text{mol.m}^{-2}.\text{s}^{-1}$ ). The Eulerian budget is calculated with the Meso-NH model, the Observed surface flux budget is deduced from the area-average CO<sub>2</sub> surface flux measurements (Table 1) and the Lagrangian budget (LAG<sub>obs</sub>) is deduced from the aircraft vertical profile measurements during experiment. Each budget is averaged over the Piper and the Dimona sub-domains.

	Time of integration	Eulerian budget MODEL	Observed surface fluxes $\sum_i^{\text{obs}} C_i w' \text{CO}_{2i}$	LAG <sub>obs</sub> $w' \text{CO}_{2\text{LAG}}$
DIMONA	3.25 h	−11.5	−12.1	−16.8
PIPER	8.5 h	−10.4	−9.4	−8.6

In these conditions, the CO<sub>2</sub> surface flux can be approximated by integrating the variation of the CO<sub>2</sub> vertical profiles in the ABL between the morning and the afternoon measurements.

The vertical profiles of observed CO<sub>2</sub> concentration displayed in Fig. 10 for the 2 aircraft, are integrated up to the top of the ABL.

For the Dimona measurements (Fig. 10a), we assumed, that the CO<sub>2</sub> concentration is well mixed in the ABL. The mean depletion of CO<sub>2</sub> in the ABL between 10:00 and 13:00 UTC is almost 3 ppm. The integration up to 1600 m gives: LAG<sub>obs</sub>=−16.8  $\mu\text{mol.m}^{-2}.\text{s}^{-1}$ .

The Piper-Aztec measurements (Fig. 10b) show a well mixed layer in the morning and a larger depletion of more than 5 ppm over the nine hours of the experiment. The observations near the surface for the afternoon flight are interpolated. The integration of the variation between the two vertical profiles, up to 2000 m gives: LAG<sub>obs</sub>=−8.6  $\mu\text{mol.m}^{-2}.\text{s}^{-1}$ .

#### 4 Discussion

Three methods to establish a regional CO<sub>2</sub> budget have been tested using observations and model output analysis. Table 2 summarizes the results obtained by these methods for two sub-regions.

The CO<sub>2</sub> surface fluxes obtained by the three methods vary between 10 and 16  $\mu\text{mol.m}^{-2}.\text{s}^{-1}$ , values lower than the fluxes estimated by Gibert et al. (2007), over a region dominated by winter crops in May, and by Schmitgen et al. (2004), in the Landes in June, which were 20 and 16  $\mu\text{mol.m}^{-2}.\text{s}^{-1}$ , respectively.

The budgets calculated according to the Dimona measurements over its sub-domain are higher than the Piper-Aztec ones, using all 3 methods. The main reason is that the time of integration is shorter and centered over the maximum of assimilation for the Dimona box. Moreover, this sub-domain is smaller and mainly covered by the pine forest (68%). For this ecosystem, the assimilation rate is high at this stage of the year.

The averaged surface fluxes obtained from surface observations and from the meso-scale model are in good agreement for the two boxes. This confirms the good calibration of the ISBA-A-gs surface scheme and the reasonable quality of the meso-scale circulations representation in the model. The Eulerian model and the averaged surface fluxes methods are also in good agreement, probably because both methods share common use of surface characteristics. The two methods use the same vegetation map and the same fraction of vegetation type. It is difficult to know to what extent the good agreement between the 2 methods is related to this aspect.

In general, the model is able to reproduce the surface fluxes and the CO<sub>2</sub> concentration observed by the aircraft. The high level of realism of the Meso-NH simulation gives confidence to the estimation of the Eulerian budget. Indeed, the Meso-NH budget calculation shows that the contribution of carbon assimilation by the vegetation is higher than the advection of CO<sub>2</sub> into the domain in both aircraft boxes, causing the diurnal CO<sub>2</sub> decrease in the ABL.

The interpretation of the results obtained with the Lagrangian aircraft budgeting is more complex. For the Dimona box, the Lagrangian budget leads to a higher surface flux than the two other methods. The reverse result is obtained in the Piper-Aztec box. Various explanations can be proposed:

- First, while the morning profiles of CO<sub>2</sub> flown over the sea are comparable for the two aircraft, the afternoon ones are incomplete in both cases, particularly in the lowest layer (see green lines in Fig. 10). The two inland profiles have been extrapolated in the lowest layers. This could introduce uncertainties in the estimation of the Lagrangian term.
- Secondly, although we tried as much as possible to plan the afternoon aircraft flights in order to follow and to sample the same air mass as during the morning flight, following wind observations, uncertainties cannot be avoided. For instance, variations of the wind field within the meso-scale domain are difficult to take into account in the forecast of the air mass trajectory.

These sources of errors will now be discussed in order to conclude the discussion about the respective advantages of each method.

First, for the Lagrangian experiment, the estimation of regional CO<sub>2</sub> surface flux from aircraft observations is difficult due to uncertainties linked to the strategy of sampling itself as mentioned above. In our case, probable errors are due to the technique of measurement of CO<sub>2</sub> vertical profiles and to the difficulties in monitoring the evolution of meteorological conditions over several hours. This last point generates uncertainties in the determination of the localisation of the air mass. The air mass sampled in the afternoon by the two aircrafts is not located at the exact place as expected and the resulting error is more than 30%, at least for the Dimona.

We deduce from Eq. (2) that the Lagrangian method uncertainty is:

$$\frac{\Delta \text{LAG}}{\text{LAG}} = \frac{\Delta \text{CO}_2}{\text{CO}_2} + \frac{\Delta z}{z} + \frac{\Delta t}{t}$$

Assuming there is no error in the altitude  $z$  during the integration,  $\frac{\Delta z}{z} = 0$ .

For the Dimona, the error in the CO<sub>2</sub> concentration measurement ( $\pm 0.3$  ppm) is  $\frac{\Delta \text{CO}_2}{\text{CO}_2} = 10\%$ . The main error is linked to the sampling strategy, because in that case the distance between the morning and the afternoon vertical profiles is almost 73 km, and the mean wind measured by the aircraft is  $4.8 \text{ m.s}^{-1}$ . According to these values, the afternoon vertical profile should have been measured 4.25 h after the morning profile, in order to sample the same air mass, instead of the effective 3.21 h. So, the error made on the time is  $\frac{\Delta t}{t} = 32\%$ .

The total uncertainty for the Dimona flights is thus almost 42%. For the Piper-Aztec, the CO<sub>2</sub> concentration measurement error is  $\pm 0.2$  ppm and  $\frac{\Delta \text{CO}_2}{\text{CO}_2} = 3\%$ . Here again, the main uncertainty is linked to the strategy of sampling the air mass. Considering 163 km between the two vertical profiles and an air mass displacement at  $4.5 \text{ m.s}^{-1}$ , the mean wind speed measured along the drifting balloon trajectory, the time between the two vertical profiles should have been 10 h instead of the effective 8.5 h, that is to say 18% error arising from the time of integration. The total error for the Piper-Aztec flights is thus almost 21%. On average across the two flights, the error is approximately 31%.

The budget calculated with the upscaling of the observed surface fluxes ( $\sum_i^{\text{obs}} C_i w' \text{CO}_{2i}'$ ) includes an uncertainty due to measurements by the eddy correlation system, which is currently estimated at around 10% (from Richardson and Hollinger (2007) method). For example, Béziat et al. (2008) estimate the error in CO<sub>2</sub> flux measurements to be approximately 7% for the rapeseed site, Auradé and 12% for the wheat site, Lamasquère in 2005. Furthermore, we have to take into account two additional sources of uncertainty linked to this method:

- first, we have to consider the uncertainty due to the regional variability of the CO<sub>2</sub> fluxes for a type of ecosystem. For instance, for a crop like maize, there is a difference in CO<sub>2</sub> flux between the irrigated and non-irrigated fields as well as in effects, depending on the agricultural practices, on the micro-climate. From the CERES observations, we calculate that this variability could lead to almost 18% of error in the diurnal net CO<sub>2</sub> flux, for the two documented fields of maize.
- secondly, another source of uncertainties concerns the estimation of the fraction of each landcover type in the given domain. In order to estimate this last source of error, the Ecoclimap database has been compared with data from “AGRESTE”, a French departmental

agricultural database (by CNRM, Météo France, personal communication, 2008). This comparison shows a difference of around 15% between the databases (according to S. Faroux, private communication, 2008).

In sum, the error in upscaling surface fluxes can reach approximately 43% to 50%, which is higher than the error for the Lagrangian estimate.

The errors associated with the meso-scale Eulerian budget are also difficult to estimate. Indeed, for this estimation, a model statistic over a long period would be necessary to estimate the model error on the wind and the CO<sub>2</sub> concentration in the ABL. Of course, such a statistic cannot be computed for a single day of simulation. Nevertheless, as discussed before, the surface fluxes of CO<sub>2</sub>, latent and sensible heat, as well as the atmospheric variables such as temperature or CO<sub>2</sub> concentration have been compared with observations and validated. Moreover, the high coherence in the model between all the terms (advection vs vertical diffusion) give confidence in the model budgeting approach.

## 5 Conclusions

This paper describes several methods to determine a regional budget of CO<sub>2</sub> using in-situ observations as well as numerical tools. The paper also includes a validation of a meso-scale model able to calculate the regional CO<sub>2</sub> budget, thanks to the full interaction of water and carbon exchanges between the surface and the atmosphere.

The observations used in this study come from a Lagrangian experiment conducted during the CERES campaign over a coniferous forest, South-West of France and simulated with a meso-scale meteorological model, Meso-NH.

This Lagrangian experiment took place during a westerly wind episode, when an oceanic air mass was sampled in the morning, near the Atlantic ocean coast line and sampled again further inland several hours later. This experiment was conducted twice, by two different research aircraft. For the Dimona aircraft, between the morning and the afternoon measurements, the air mass CO<sub>2</sub> concentration decreased by almost 3 ppm over 3.25 h. For the Piper-Aztec aircraft, the decrease reached 5 to 6 ppm over 8.5 h.

The meso-scale model was first evaluated against radiosounding, CO<sub>2</sub> surface flux observations, CO<sub>2</sub> measurements at 2 sites and CO<sub>2</sub> concentration from the aircraft observations. The comparisons showed that the model is able to reproduce correctly both the ABL development and the CO<sub>2</sub> mixing ratio in and above the ABL as well as the NEE.

Next, three methods were used to determine the regional CO<sub>2</sub> surface flux: a numerical model budget calculation using an Eulerian method, upscaling of surface fluxes observations and Lagrangian observations by the two aircraft.

All of the budgets are in reasonable agreement as displayed in Table 2. The regional surface flux is between  $-10$  and  $-16 \mu\text{mol.m}^{-2}.\text{s}^{-1}$  depending on the time of integration

and the areas considered. More specifically, the estimations of the regional CO<sub>2</sub> surface flux by the Eulerian meso-scale model budgeting and by the observations at the surface flux sites are very similar.

The authors tried to determine as much as possible the main sources of error for each of the 3 methods. For the estimate taken from the upscaling of surface fluxes upscaling, in addition to the measurement errors, we have to consider errors caused by the assumption of homogeneity for a given ecosystem everywhere in the domain. Another source of error is related to the accuracy of the areal occupied by each ecosystem. As a result, the surface flux upscaling method, should be most useful for a relatively homogeneous domain. In our case, despite the large number of flux stations (8 for a domain of  $200 \times 200 \text{ km}$ ), it would have been easier to estimate the regional flux with at least 4 more stations, especially for the larger PIPER Box. In CERES, a number of landcover types were not available, including deciduous forest, bare ground, and several winter or summer crops.

For all of these reasons, the Lagrangian, which is an integrated measurement over space and time, is more efficient than the upscaling of surface fluxes, although the experimental strategy is difficult to operate (due to, for example, gaps in data, localisation of the air mass).

The authors would like to stress the necessity for future Lagrangian campaigns to pay particular attention to the vertical profiles, especially in the lower part of the ABL, in order to reduce the associated uncertainties.

The meso-scale model gives an approximate value for the regional CO<sub>2</sub> budget. We cannot attribute an error estimation to the model because of a lack of statistics. However, the model coherence and the comparisons with the observations are good enough to have a relative confidence in the modeling approach.

As a conclusion, the atmospheric CO<sub>2</sub> plays a key role in the changing climate but the underlying processes, particularly the surface-atmosphere interactions, induce difficulties in quantifying the budget, even at the regional scale. This study, based on both observations and numerical tools, tries to estimate as accurately as possible this budget, assessing the associated uncertainties. The authors give some way to reduce the sources of error, for future experimental campaigns. Concerning the improvement of the numerical method, the next step would be to associate an inverse modeling at the regional scale, using the modelled meteorological and concentration fields in order to retrieve the CO<sub>2</sub> sources and sinks accurately. Lauvaux et al. (2008a,b) have already showed the high potential of the Meso-NH model associated with a Lagrangian Particle Dispersion Model (LPDM), to estimate sources and sinks of CO<sub>2</sub>.

The CERES database offers the opportunity to perform a similar study for a large number of cases with different environmental conditions encountered during the Spring and Autumn 2007 campaigns, for which these methods could be improved and the CO<sub>2</sub> budget improved.



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