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THE EXCHANGE OF CARBON DIOXIDE BETWEEN THE ATMOSPHERIC BOUNDARY LAYER AND THE FREE ATMOSPHERE: OBSERVATIONAL AND LES STUDY

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1. INTRODUCTION

Previous studies on carbon dioxide (CO_2) diurnal variability focus mainly on surface processes such as photosynthesis and respiration, whereas little is known about the exchange of CO_2 between the atmospheric boundary layer (ABL) and the free atmosphere (FA). This last process is closely related to the entrainment of heat at the interface. Vilà et al. (2004) studied this process in a situation observed at Cabauw in the Netherlands. They analyzed the role of the boundary layer evolution on the diurnal variability of CO_2, focusing in particular on the exchange of CO_2. Entrainment appeared to be an important aspect in the evolution of CO_2 in the morning hours, however, in the afternoon it becomes less relevant and surface processes are the dominant factors.

Our research extends the previous study by analyzing two convective boundary layers, the first observed at Cabauw in the Netherlands (RECAB) and the second near Beaumont in the USA (CASES-97) (LeMone et al., 2000, 2002). Surface and upper air measurements from these sites are analyzed and combined with Large-Eddy simulation results to study the evolution of the CO_2 mean and the vertical flux profile. Furthermore, the evolution of the potential temperature and the entrainment of heat are also studied as these are fundamental for the development of the boundary layer (Garratt, 1992; Stull, 1988).

Sensitivity analysis is conducted to determine which factors play an important role in the exchange of CO_2 and therefore in the CO_2 diurnal variability.

2. METHODS

2.1 *OBSERVATIONS*

In RECAB, spiral vertical profiles and horizontal legs were flown to measure at high frequency three-dimensional wind, temperature, specific humidity and CO_2 concentrations above Cabauw on 27 July 2002. In order to obtain reliable statistics and spectra for the turbulence measurements three or four passages of approximately 8.5 km were flown successively at a constant height in the morning (8 UTC) and in the afternoon (13 UTC). A cloudless sky was observed until 14 UTC and the wind speed was 2-3 ms^{-1}.

The CASES-97 field project took place in the Walnut River Watershed in Southeast Kansas between 21 April to 21 May 1997 (LeMone et al., 2000, 2002). The University of Wyoming King Air flew horizontal legs of approximately 45 km near Beaumont on 10 May in the morning (9-12 CST). We split the horizontal legs of Beaumont into three parts, 15 km each, in order to be able to retrieve more robust statistics and to calculate standard deviations. The meteorological conditions were characterized by clear sky and the wind was steady with a windspeed of 5-6 ms^{-1}. There was strong subsidence and the water mixing ratio above the boundary layer was extremely low (near zero).

Both sites were characterized by grass vegetation, although the grass vegetation uptake of CO_2 was larger at Cabauw. In the early morning at 6.30 CST, a radiosonde at Beaumont showed a stable boundary layer and a residual layer above it, whereas the vertical potential temperature morning profile measured by the aircraft above Cabauw does not show a residual layer and appeared to be well mixed at 8 UTC. The potential temperature difference between the ABL and the FA is 2.5 K for Cabauw and 2 K for Beaumont. The CO_2 difference is -30 ppm for Cabauw and -0.5 ppm for Beaumont. In the two situations, we assume that the shear contributions at the surface and at the entrainment zone was negligible compared to convective turbulence.

2.2 *SETUP NUMERICAL EXPERIMENTS*

In order to complete the understanding of these two convective boundary layers, a Large-Eddy Simulation (LES) model is used in this study. In this

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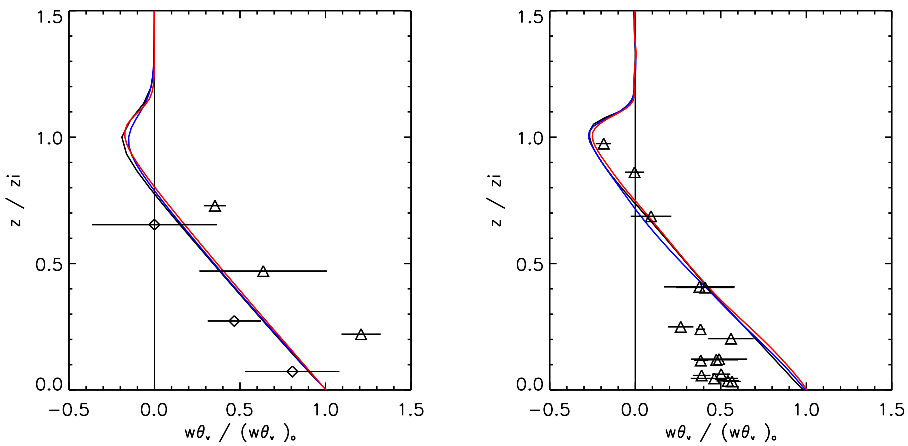

FIG. 1: *Vertical LES profiles of dimensionless buoyancy fluxes at Cabauw (left) and at Beaumont (right). The bars represent the standard deviations from the observations. A triangle represents observations in the morning and a rhombus represents afternoon observations. The continuous lines are the LES results for 8 LT (black line), 10 LT (blue line) and 12 LT (red line).*

model, the surface fluxes and the initial profiles of potential temperature, moisture and carbon dioxide have to be prescribed. The prescribed surface fluxes are sinusoidal functions based on the measured surface values. The initial profiles are similar to the ones observed by the aircrafts at Cabauw and by the radiosonde at Beaumont. The considered domain is 6.4 km x 6.4 km and the prescribed resolution is 100 m for the horizontal in both directions and 25 m for the vertical. More details about this model can be found in Cuijpers and Duynkerke (1993) and Cuijpers and Holtslag (1998).

3. RESULTS AND DISCUSSION

The exchange of potential temperature and carbon dioxide is studied by combining observations and LES modeling. Two convective boundary layers, at Cabauw and at Beaumont, are analyzed.

The vertical LES profiles of the dimensionless buoyancy fluxes and the observations with their respective standard deviations for Cabauw and Beaumont are shown in Figure 1. The fluxes are scaled by the measured surface values. For Cabauw, the LES results lie inbetween the observations. For Beaumont, the observations near the surface are too low, but higher in the boundary layer the values agree considerably well. The flux calculations based on 15 km horizontal legs yield similar results to the ones discussed by LeMone et al. (2000, 2002). The dimensionless buoyancy fluxes from the LES model show a similar pattern for both cases. The ratio between the entrainment flux and the surface flux shows how important the entrainment process is with regard to the surface process. This ratio is -0.2 at Cabauw and -0.3 at Beaumont, which agree with previous numerical studies (Pino et al., 2003; Van Zanten et al., 1998).

The vertical LES profiles of the dimensionless CO_2 fluxes and the observations are shown in Figure 2. Again, the fluxes are scaled by the measured surface values. As is noticed from the figures, there is larger scatter between LES and observations for CO_2 than for heat. Moreover, the scaling of the fluxes does not provide the coincidence of the LES profiles, which can be probably explained by the fact that the CO_2 exchange process and the surface process are independant processes. The calculated fluxes from the observations show a lot of scatter, in particular at Beaumont, making it difficult to find a comprehensive behaviour of the evolution of the vertical flux profiles. This indicates a larger uncertainty in CO_2 measurements and therefore this research is mainly focused on the LES results with the observations as a supportive aspect. The CO_2 exchange fluxes from the LES model show different regimes, namely positive exchange fluxes at Cabauw and negative exchange fluxes at Beaumont. The exchange ratio decreases in time from 1.7 to 0.2 at Cabauw, whereas at Beaumont this ratio decreases from 0 to -0.3.

Although the temperature exchange show a comparable situation at both sites, the CO_2 exchanges are different. The positive CO_2 exchange dilutes the CO_2 concentration in the convective boundary layer, whereas the negative exchange flux enhances the CO_2 concentration. In addition, the larger the difference in CO_2 concentration is

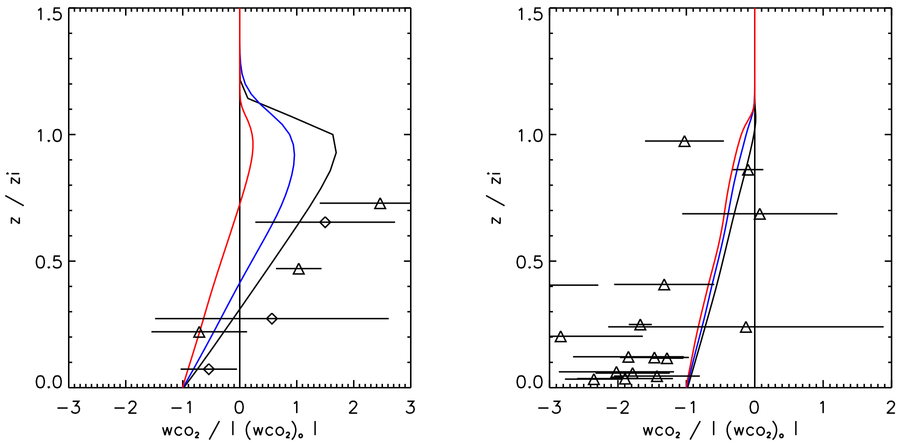

FIG. 2: *Vertical LES profiles of dimensionless carbon dioxide fluxes at Cabauw (left) and at Beaumont (right).*

between the ABL and the FA in the early morning, the more important the CO_2 exchange becomes. In conclusion, the Cabauw results support the conclusion from Vilà et al. (2004) that the CO_2 exchange process is most important in the morning hours. However, the Beaumont results reveal that the CO_2 exchange is becoming more important in the early afternoon.

An important aspect of our research is to determine the role of the boundary layer dynamics in the CO_2 diurnal variability under convective conditions. Numerical experiments have been carried out to determine the importance of boundary layer growth and initial concentrations on the diurnal CO_2 variability.

We first focus on the role of the mean vertical velocity, namely subsidence, in the CO_2 exchange. It should be noticed that the vertical advective flux due to subsidence can be an additional value (positive or negative) to the exchange of CO_2. A simple zero-order mixed layer model (Betts, 1992; Carson, 1973; Tennekes, 1973) can illustrate this relation If we assume that the CO_2 exchange flux is a function of the entrainment velocity (w_e) and the CO_2 concentration jump ($\Delta CO_2 = CO_2(FA) - CO_2(ABL)$), then we can write

$$\overline{wCO_{2e}} = -w_e \Delta CO_2, \quad (1)$$

where the entrainment velocity is defined as

$$w_e = \frac{\partial h}{\partial t} - w_s. \quad (2)$$

In the right hand side, $\frac{\partial h}{\partial t}$ represents the boundary layer growth ($\frac{\partial h}{\partial t} > 0$) and w_s is the subsidence velocity ($w_s < 0$).

Two numerical LES experiments were set up to study the role of subsidence: (a) $w_s = 0$ and (b) $w_s = -0.01 m s^{-1}$, constant with height and time. In Figure 3, the time evolution of the potential temperature and the CO_2 concentration calculated by the LES model without and with subsidence is shown. The inclusion of subsidence in the simulations has the following consequences: (1) the boundary layer becomes shallower (nearly 200 m less, not shown here), (2) the boundary layer becomes warmer due to the additional vertical advection of heat and (3) the dilution of CO_2 becomes more effective. The simulations show that the constant presence of subsidence is a significant factor in the dilution of CO_2.

Briefly, in the morning hours the large initial negative CO_2 jump, due to nocturnal respiration, diminishes rapidly (approximately 5 ppm per hour) on account of the large positive exchange flux. Then, this large positive exchange flux is reduced by a decreased CO_2 jump during the development of the boundary layer. The additional presence of subsidence will cause a faster dilution of the CO_2 concentration in the ABL compared to the simulations without subsidence. At a certain moment, the same CO_2 value as in the free atmosphere (362 ppm) is reached and even continues to decrease below this value due to the surface process, which is large during the hours of maximum solar radiation. This will cause negative exchange fluxes in the afternoon due to a positive CO_2 jump ($\Delta CO_2 > 0$).

In the near future, we will elaborate on this study by introducing horizontal advection. Besides this, other days in CASES-97 or in other field projects will be analyzed to support our findings.

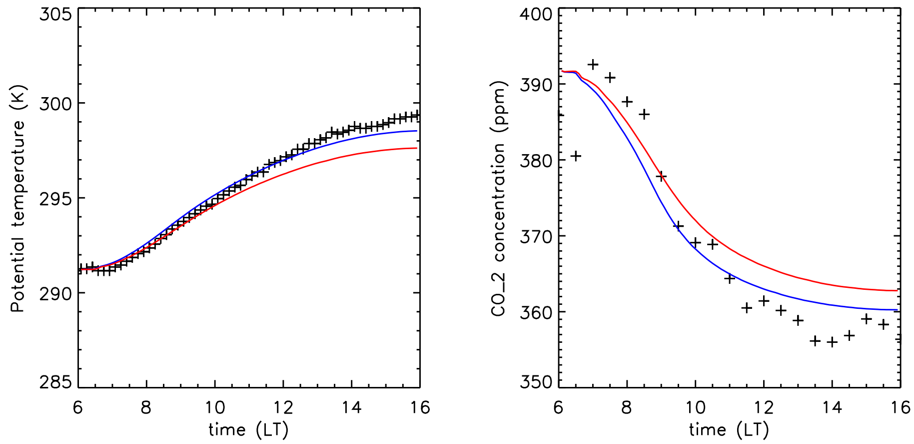

FIG. 3: *Evolution of the potential temperature (left) and the carbon dioxide concentration (right) at Cabauw calculated by the LES model without subsidence (red line) and with subsidence (blue line). The results are compared with the Cabauw tower observations measured at 200 m (plusses).*

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