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1. INTRODUCTION

Carbon cycle research has gained prominence in recent years due to its established links with global climate change. One critical component of carbon cycle research is estimating the annual transfer of CO₂ between the atmosphere and the biosphere (e.g., plants, soils). A global network of over 140 towers has emerged over the last 5-10 years with the common goal of measuring this transfer of CO₂ over a variety of ecosystems worldwide. Each uses similar techniques (e.g., eddy site covariance and storage methods) to measure fluxes of CO₂. There exists a third component of the total CO₂ transfer, the advective flux, which is often assumed negligible. Advection can be caused by heterogeneity of the source areas (Yi et al., 2000), or by large-scale changes in the atmospheric concentration of CO2 that are advected past the tower over periods of seconds to days (Hurwitz et al., 2004).

This study focuses on measuring the advective component of carbon dioxide transfer at a relatively ideal site, the Cabauw tall tower in the Netherlands, during two months (September, 2003 and April, 2004). The tower is located in flat and homogenous grassland terrain. Flux measurements are made at four levels on the tall tower, allowing the advective flux to be estimated by measuring the difference in Net ecosystem exchange (NEE) of CO₂ between This manuscript focuses on diurnal levels. averages to assess persistent changes in the advective flux. Ongoing research not presented here includes implementing strategies to separate the most likely components of advection through footprint analysis.

2. STUDY SITE AND MEASUREMENTS

2.1 The Cabauw Measurement Site

The Cabauw meteorological tower was established in 1973 specifically for studying the atmospheric boundary layer (Van Ulden and Wieringa, 1995). The tower (213 m) is located in the center of Netherlands (51°58'N, 4°56'E), approximately 50 km E-SE of the North Sea and 1 km NW of the River Lek. The site was chosen due to the representativeness of the surrounding landscape, and because at that time no further urban agglomerations were planned within 15 km of the site. The immediate region surrounding the tower has been described in detail previously (e.g., Van Ulden and Wieringa, 1995; Beljaars and Bosveld, 1996 and references therein) and consists primarily of grassy meadows and ditches; one village exists to the east ~ 1.5 km from Cabauw (Figure 1). The changes in surface elevation are at most a few meters over the surrounding 20 km, and ~400 meters of open pastures exist in all directions around the tower. To the West, the meadows extend for several kilometers, and to the East meadows give way to trees and low buildings for the next several kilometers (Beljaars, 1982). At distances greater than 3 km the dominant land type is grass; however, other land types such as crops and trees increase in importance. The vegetation cover at Cabauw and regionally around the tower is nearly 100% throughout the year with the dominant grass species consisting of *Lolium perenne*. Lesser amounts of Poa trivialis and Alopecurus geniculatus exist in the surrounding region (Beljaars and Bosveld, 1996).

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Figure 1. The LGN4 land-use (25-m) classification surrounding the Cabauw tall tower (located at the cross hairs in the middle). Green is grass, blue is water, red indicates city regions, black dots are tree groups.

2.2 Methods

Vertical profile measurements of CO₂ were maintained by ECN at 20, 60, 120, and 200-m heights during the period of this study. CO₂ concentrations were measured using a Siemens Ultramat 5 NDIR with a resolution of 0.1 ppm. The integration time was 1 minute, and the switching time between the levels was 2 minutes. CO₂ concentration measurements along the tower were made in a cycle of 8 minutes, the concentration profiles were averaged to 30 minute intervals. Daily zero and span calibrations were carried out against working standards, and these standards were calibrated monthly against NOAA CMDL certified calibration gases. Air was dried at the inlets to about 15 degrees lower than the dewpoints of ambient level using Permapure dryers. As it is quite common that the top of the boundary layer is below the highest level of the tower at night, there is often the opportunity to measure the storage flux (F_{ST}) accurately during low-turbulence conditions.

Turbulent fluxes were measured in-situ at each level with open-path analyzers at the 5-m, 60-m, 100-m and 180-m levels. At the 5-m level, an IFM open-path gas analyzer (Kohsiek, 2000) was used for turbulent measurements on a tower ~ 200 m from the base of the Cabauw tower since 2001. Turbulent CO_2 flux data from the IFM was compared with data collected using a LI-COR LI-7500 open-path CO_2 analyzer and showed that the IFM underestimated the flux by ~ 17 % compared to the LICOR. From 2003-2005 Cabauw was instrumented with additional LI-7500 infrared gas analyzers for measurement of turbulent CO_2 fluxes at 60, 100, and 180-m heights. These data have been made available for this and other projects within the CarboEurope and CESAR programs.

All flux sensors were sampled at 10 Hz, and raw data was archived for further analysis. Other meteorological sensors on the mast were sampled at 3-s or 12-s intervals. Statistics including averages, variances, covariances, maximum and minimum values are automatically computed on 10-minute intervals and transmitted to the meteorological service (KNMI) in de Bilt. Near real-time 10-minute averages were plotted and the KNMI web displayed on site (www.knmi.nl/~bosveld) for control purposes. The archived 10-minute data is manually inspected for suspect data (typically recorded during periods of rain or dew formation). Suspect data points are removed from the data and the 'cleaned' data are stored in new 10-min files. Ten-minute samples are averaged and summed with the low-frequency contribution for the period of final averaging (30 or 60-minute averages).

The scalar conservation equation is used to mathematically describe the three terms contributing to NEE: F_{EC} , F_{ST} , and F_{AD} . Starting with the scalar conservation equation in the typical case were the horizontal scale of the flow field is much larger than the depth of the boundary layer, NEE can be estimated (Yi et al., 2000) by

$$NEE = \int_{0}^{Zr} \frac{\partial \overline{c}}{\partial t} dz + (\overline{w'c'})_{Zr} + \int_{0}^{Zr} \left\{ \overline{u} \frac{\partial \overline{c}}{\partial x} + \overline{w} \frac{\partial \overline{c}}{\partial z} \right\} dz$$
$$NEE = F_{ST} + F_{EC} + F_{AD} \qquad [1].$$

Here mean quantities are expressed with an overbar and are averaged over the measurement period, and prime values indicate instantaneous deviations from the mean quantities. The first term on the right-hand side of [1] is the storage flux (F_{ST}) , and is calculated as the change in the mean CO_2 concentration profile with time $(\partial c/\partial t)$, integrated over height (z) to the top measurement height on the tower (Z_r) . The second term is the turbulent flux of CO₂ (F_{EC}), calculated as the timeaveraged product of the turbulent components of vertical wind velocity (w) and CO₂ density (c) at the measurement height (Z_r) above the ground (Baldocchi et al., 1988; Lenschow et al, 1995). The final term in [1] is the total advective flux of CO_2 , which includes a horizontal component and a vertical component. The total advective flux can be measured as the difference in the sum of F_{EC}

and F_{ST} between two measurement levels, as demonstrated at the WLEF tall tower (396 m), an AmeriFlux site in N. Wisconsin, U.S.A. (Yi et al., 2000).

3. RESULTS

3.1 Diurnal Cycle of CO₂ Concentration

The monthly average of half-hourly concentrations of CO₂ are plotted in Figure 2 for September, 2003 and April, 2004 for the levels at which eddy covariance measurements were Where the measurements were not made. available (e.g., 5-, 100-, and 180-m levels) the CO₂ concentration was either modeled using tested flux-profile relationships (5-m level) or scaled between heights based on the measurements above and below the EC measurement height. The modeling of the 5-m layer was verified based on a period in the 1990's when both 20- and 5-m measurements were available.

On low-wind nights, when a stable boundary layer forms, the CO₂ concentration increases near the Earth's surface causing



Figure 2. Diurnal cycle of the CO_2 mixing ratio for September, 2003 (top) and April, 2004 (bottom). Data presented is the average of all half-hourly values.

higher concentrations to be recorded at the lowest levels of the tower. The 5-m level was modeled to be particularly variable during night-time periods for both months, however this could be result of the modeling process. All levels except for the 180-m level showed some degree of increasing nighttime periods; CO₂ during the CO_2 concentration at 180-m was relatively stable between day and nighttime periods during both months. During the daytime hours the boundary layer is mixed due to convection causing very small differences among levels on the tower. The period of mixed-gradient conditions typically began at 0900 local standard time and ended between 1700 and 2000. The range of the CO₂ gradient (minimum daytime measurements to maximum nighttime) was much larger during September than during April (~ 90 ppm and 50 ppm, respectively); however, the daytime mixed values of CO₂ for April were ~ 10 ppm higher than those observed in September.

3.2 Diurnal Cycle of CO₂ Fluxes

In Figure 3 the monthly average of halfhourly values of turbulent flux, storage flux, and NEE are plotted for September, 2003 and April, 2004. During nighttime periods there was enough turbulence at the 5 and 60-m levels to record turbulent fluxes; this turbulence was absent from the higher levels resulting in near-zero values until 0700 LST. In September late-night/ earlymorning turbulent fluxes displayed persistent increases for all levels which was absent during the April measurements. The observed increase in turbulent flux occurred progressively later with height on the tower. Specifically the increase started as early as 0300 for the 5-m level, 0500 for the 60-m, and 0700 for the 100-m and 180-m levels.

Storage fluxes for both months show very similar patterns with strong negative averages in the morning transition period and largely positive, values during night-time periods. Storage fluxes were near-zero in value between 1400 and 1600 before becoming positive again. The minimum storage flux at each height lagged minutes behind the next lower level during the morning transition. In September the value of storage flux at the higher levels was nearly three times the maximum uptake observed by the 5-m level during daytime hours, and in April, the value of storage flux was similar in magnitude to daytime uptake.

The graphs of NEE are similar in that both show relatively no difference in NEE between the measurement heights between ~ 1100 and 1700



Figure 3. Monthly average of the half-hourly values of turbulent and storage flux (as mg CO₂ m⁻² s⁻¹), and NEE for the Cabauw tower during September, 2003 (left) and April, 2004 (right).

local standard time. However, in September there exists a strong and persistent difference in NEE between each level from ~0500 until 1100 with each measurement height displaying first positive values of NEE at the beginning of the morning transition and then negative values, similar in magnitude to the storage fluxes, near the end of the morning transition between 0800 and 1000. In April, NEE is markedly similar between the 60, 100, and 180-m levels during all hours of the day, and different for the 5-m level. The 5-m level displays an earlier drop in NEE starting at 0400, whereas the other levels start to drop following 0500. Again, as in September, minima in NEE were observed between 0800 and 1000 and were strongly influenced by the magnitude of the storage flux.

Persistent advective fluxes, as observed by the difference in NEE (Δ NEE) between levels (Figure 4), were observed at Cabauw associated with the morning transition period during



Figure 4. Monthly average of the half-hourly values of \triangle NEE for the Cabauw tower during September, 2003 (left) and April, 2004 (right). 'D5-60 NEE' above represents \triangle NEE between the 5-m level and that of the 60-m level.

September, 2003 for both the 5-60-m and the 60-100-m intervals. There existed a different pattern for Δ NEE in April than in September; Δ NEE was less pronounced but lasted the majority of the daytime hours. The magnitude of Δ NEE between the 5 and 60-m levels and 60-and 100-m levels was stronger in September than in April, reaching nearly 80% of the magnitude of NEE in September at the 100-m level. In both months the Δ NEE observed between the 100- and 180-m levels was rarely significantly different from zero.

4. DISCUSSION AND CONCLUSIONS

4.1 Diurnal Cycle of CO₂ Concentrations and Fluxes

The diurnal cycle of atmospheric concentrations and fluxes were generally what would be expected based on previous data published in the literature. Daytime uptake during both months was typical of previous measurements at Cabauw during years of non-drought conditions (Hensen et al., 1997). The

data from September, 2003, however, was particularly interesting with peculiar increases of turbulent flux in the early morning periods that have not been observed in the past. The following discussion focuses on these measurements, their causes, and the effect on the diurnal cycle of advective flux.

Closer inspection of the individual turbulent fluxes throughout September showed that 15 days were affected by the increases of turbulent flux (based on the 60-m level) during early morning hours. At the 60-m level these increases occurred generally between 0500 and 1100, and typically between a narrower window of 0800-1000 LST.

The CO_2 stored in the lower boundary layer during stable nights is transported to higher levels as the mixed layer grows. In both months the high values of positive turbulent flux occurred primarily during morning periods when the specific storage flux was negative (Figure 5c.); this was most evident in September. Negative storage fluxes occur when the previous value of storage (i.e., the concentration times the height) is higher than the present measurement, indicating the CO₂ is being exported or advected from the lower layers to higher layers. While the vertical concentration gradient exists, the CO₂ near the ground is higher than in the layer above, resulting in a positive turbulent flux. Thus, peaks in the fluxes and CO₂ profiles consecutively reach higher levels as the mixed layer grows to eventually reach a maximum height by late morning. As will be discussed below, the magnitude of the positive and increasing turbulent fluxes during certain days of September was likely the result of the formation of strong concentration gradients at night due to: (1) contribution of CO₂ from anthropogenic sources from certain wind sectors, and (2) occasional low-turbulence conditions. Strong gradients were not thought to result from increased respiration rates.

While average temperatures during September, 2003 were about 5°C higher relative to April, 2004, which could have resulted in increased respiration rates, we observed that average 5-m turbulent fluxes during nighttime periods were essentially the same for September and April (~0.1 mg m⁻² s⁻¹). These values are at the lower limit of what has been observed for respiration at Cabauw in the past during these months (Hensen et al., 1997). For instance, for years 1993-96 the average nighttime value of respiration varied between 0.1 and 0.3 mg $CO_2 \text{ m}^{-2} \text{ s}^{-1}$ in both September and April, with maximum values up to 0.6 mg CO₂ m s⁻¹ (Hensen et al., 1997). In this study, nighttime respiration was lowest (0.1 mg CO₂ m s⁻¹) for 1996 and was explained by drought conditions. As 2003 was a particularly dry year, we expect that drought conditions likely reduced the respiration potential in September, and did not lead to excessive build-up of CO₂ from biogenic sources during nighttime periods.

Higher atmospheric concentrations of CO₂ could, however, be related to anthropogenic contributions from populated regions like the village of Lopik to the E, Ameide to the SE, and Langerak to the SW, or transport from further industrial centers south of Cabauw (e.g., Figure 5a. shows that CO₂ Rotterdam). concentrations increased dramatically between wind directions of 75 and 90 degrees for the 60height, especially during September. m Concentrations remained high between 90 and 270 degrees, and reduced again in the W-NW sector. Daytime and nighttime concentrations were higher when the wind was from the 90-270 sector indicating likely contributions from anthropogenic sources in this upwind region. The periods with increasing, and strongly positive, fluxes occurred when the wind direction was from directions where higher atmospheric concentrations of CO₂ were observed (Figure 5a

and 5b). Both months showed higher concentrations from this sector, however the difference in concentration between the 'clean sector' (280 - 75 degrees) and the 'dirty sector' (90-270 degrees) was much less pronounced in April due to the higher background level of atmospheric CO_2 . Average atmospheric values of CO_2 in April were on the order of 10 ppm higher than September, which is consistent with the seasonal variability of CO_2 in this region (~25 ppm for Cabauw). In April the highest positive values of turbulent flux were not observed from a particular wind direction as was observed in September.

Another likely factor contributing to high build-up of CO_2 and high turbulent fluxes in morning periods is low turbulence conditions during the night. High fluxes were not observed when the wind speed was higher than 6 m s⁻¹ (at the 60-m level) and most occurred when the wind speed was $\leq 3 \text{ m s}^{-1}$ (not shown). These periods were also characterized by u- < 0.2 m s⁻¹ measured at the 5-m level. Likewise, most elevated fluxes at the 100-m level occurred when wind speeds were $\leq 3 \text{ m s}^{-1}$. This implies that buildup of CO_2 is possible when the wind speeds are generally low at the tower, thus resulting high turbulent fluxes in the morning hours related to the venting process.

4.2 Diurnal Cycle of Persistent Advective Flux

Typically the sum of turbulent and storage fluxes are considered to be a good approximation of NEE if the sources/sinks of terrain are homogeneous and the terrain is flat (Yi et al., 2000). This study suggests that even when these conditions are met, as should be the case during daytime (well-mixed) conditions at Cabauw, there still might be a significant advective flux that can be approximated by differences in NEE on different levels of a tall tower.

In this study, anomalous turbulent fluxes in September, 2003 had a noteworthy effect on the advective component of flux, especially during the morning transition through to mid-day, and at levels up to 100-m. In April, 2004 the differences in NEE between levels were not affected as much by the morning transition, but still continued to display a subtle, but significant advective flux throughout the day. Yi et al. (2000) observed that Δ NEE was largest after calm nights, which was consistent with their finding that most of the advective flux stemmed from the storage term. In September, we observed both the turbulent flux and the storage term contributing strongly to advection.



Figure 5. CO_2 concentration with wind direction (a), turbulent flux with wind direction (b), and turbulent flux plotted against storage flux for September (blue diamonds) and April (open pink diamonds). All data is for the 60-m level.

In both September and April the average hourly value of advective flux was between 0.2 and 0.4 mg CO_2 m⁻² s⁻¹, which is between 30 – 60 % of the maximum value of NEE. The diurnal integral of advective flux between the 60 and 100-m level was 57 % of the integrated NEE for the 100-m level in September, but only 6 % of the 100-m level for April. In Yi et al. (2000), the integrated value at their 122-m level was 27 % of NEE in July, and was thought to be a result of horizontal advection due to complex terrain, and because the magnitude of the storage term increases with altitude of the flux measurement. At the higher levels they also observed that the maximum advection was observed during mid-day, which they attributed solely to horizontal advection rather than vertical, given that mid-day gradients are very

small. Our results show that mid-day maximums in Δ NEE are not a persistent phenomena as these were not observed during September (Figure 4). However, the patterns of \triangle NEE for April were very similar in shape to those published by Yi et al. (2000) for July for both the low and high-height intervals, suggesting that similar atmospheric processes can lead to systematic advective patterns at various sites, regardless of the homogeneity of the terrain. Yi et al (2000) attributed \triangle NEE to significant spatial gradients in landcover. The differences observed between integrated Δ NEE from this study and that of Yi et al. (2000) (6 % and 27 % of ~100m integrated NEE, respectively), is therefore likely in part related to the influence due to landcover variability. While the terrain at Cabauw is relatively homogeneous with respect to CO_2 sources and sinks during daytime periods relative to WLEF, ongoing research of the data presented here is focused on verifying homogeneity in the source area for NEE comparisons.

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