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Atmospheric Measurement Techniques Discussions

Direct measurement of the oceanic carbon monoxide flux by eddy correlation

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Abstract

This report presents results from a field trial of ship-based air-sea flux measurements of carbon monoxide (CO) by direct eddy correlation using an infrared-laser trace gas analyzer. The analyzer utilizes Off-Axis Integrated-Cavity-Output Spectroscopy (OA-ICOS) to achieve high selectivity for CO, rapid response (10 Hz) and low noise. Over a two-day sea trial, peak daytime seawater CO concentrations were ~ 1.5 nM and wind speeds were consistently 10–12 $m s^{-1}$. A clear diel cycle in CO flux with an early afternoon maximum was observed. An analysis of flux error sources suggests air-sea CO flux measurements are best performed in regions remote from continental pollution sources.

1 Introduction

Carbon monoxide (CO) is produced in the ocean surface mixed layer by photolysis of chromophoric dissolved organic matter (CDOM) (Wilson et al., 1970; Lamontagne et al., 1971; Zuo and Jones, 1995). Daytime photolytic production and continual consumption by microbes (Conrad and Seiler, 1980, 1982) leads to a pronounced diel cycle in surface seawater CO concentration with a pre-dawn minimum and an early afternoon maximum (Lamontagne et al., 1971; Conrad et al., 1982; Johnson and Bates, 1996; Stubbins et al., 2006; Zafiriou et al., 2008). Considerable variability is possible in both the rate of production (Valentine and Zepp, 1993; Zuo and Jones, 1995) and rate of consumption (Jones, 1991; Jones and Amador, 1993; Johnson and Bates, 1996). In two detailed studies of the water column CO budget, ventilation to the atmosphere was less significant than loss to microbial consumption (Bates et al., 1995; Zafiriou et al., 2003).

Although marine emissions represent a minor fraction of the global CO budget (Bates et al., 1995; Stubbins et al., 2006), the ocean may be a significant source of CO to the remote Southern Hemisphere marine boundary layer (Erickson and Taylor, 1992). In

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addition, CO is recognized as a useful tracer for studies of sea surface mixed layer processes because it couples to biological, photochemical, and physical mixing dynamics (Najjar and Erickson III, 1995; Zafiriou et al., 2008).

Due to the lack of a suitable direct flux measurement, previous studies of oceanic CO emissions and cycling utilize empirical gas exchange formulations (e.g. Wanninkhof, 1992; Nightingale et al., 2000). CO solubility in seawater is quite low. Empirical air-sea transfer studies typically focus on gases of similarly low solubility (e.g. Rn or He/SF_6 dual tracer methods), so there is reason to believe these formulations also provide a reasonable representation of CO transfer. An eddy correlation flux measurement allows a practical test of this assumption. In a more fundamental sense, direct measurements of the CO flux facilitate development of physical gas transfer algorithms which specify the solubility dependence of the gas exchange coefficient (e.g. Fairall et al., 2011, and references therein).

In this submission we present results from a short field trial of a new method for direct measurement of the oceanic CO flux by eddy correlation. To our knowledge, this is the first reported CO flux measurement from a ship. In addition, an analysis of the CO flux data illuminates important aspects of flux measurement error for trace gases with moderate-to-long atmospheric lifetimes and high background concentrations.

2 Experimental

An LGR model 907–0014 N_2O/CO analyzer (Los Gatos Research, Inc.) was used in this trial. This instrument employs a continuous narrow-band infrared laser source for off-axis ICOS absorption measurements of N_2O, CO and H_2O (O'Keefe et al., 1999; Baer et al., 2002). A 200-tube Nafion air drier (Perma Pure PD-200T-24-SS) reduces dew point to $< -10\,^{\circ}C$, yielding essentially dry-air concentration values. The analyzer data rate is 10 Hz, but in this configuration, at a sample flow of ~ 10 std l min^{-1}, frequency response is $\sim$ 1–2 Hz. Air was subsampled from a 20 m high-flow teflon inlet (3/8″ ID) drawing air at ~ 80 std l min^{-1}. The air inlet, sonic anemometer and

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a six-channel motion sensor were mounted at the top of a 10 m meteorological tower on the bow of the University of Hawaii research vessel Kilo Moana. A precisely timed hourly injection of nitrogen at the inlet tip was used to synchronize wind and CO data. Procedures for correcting ship motion interference in sonic wind data have been described previously (Blomquist et al., 2010; Edson et al., 1998).

The mean ten-minute CO variance spectrum (Fig. 1) shows a "pink" background noise ($\sim 1/f^n$) in addition to the $-5/3$ dependency due to turbulent dissipation. The integrated noise variance over the flux bandpass (0.00167 to 2 Hz) is 0.015 ppb^2 (σ = 0.12 ppb). An instrument artifact signal is evident at 3 Hz, but this is beyond the flux frequency response and has been filtered from the data. A comparison of ship-board spectra with laboratory data shows negligible analyzer sensitivity to ship motion.

3 Results

The CO flux trial was conducted on a routine cruise to the Hawaii Ocean Time-series (HOT) station ALOHA, located at 22° 45′ N, 158° W (cruise HOT-238, 18–22 December 2011). Over two days of sampling, wind speed was consistently 10–12 m s^{-1} and relative wind direction remained within ±20° of the bow. Seawater CO concentration at 5 m depth (Fig. 2) was measured from selected daytime CTD casts over two days using the method of Xie et al. (2002). An afternoon maximum of ~1.5 nM is evident. Nighttime samples were not analyzed, but were most likely ~0.5 nM or less based on data from prior cruises, implying a mean daily concentration of <1 nM.

Seawater CO measurements at station ALOHA on prior cruises since 2008 (also Fig. 2) show a diel pattern typical for blue water regions in the Pacific and Atlantic (e.g. Johnson and Bates, 1996; Stubbins et al., 2006; Zafiriou et al., 2008): an early afternoon maximum of 2–3 nM and mean daily concentration of ~1 nM. In contrast, peak seawater CO concentrations during HOT-238 shown in Fig. 2 were about half the typical value, providing a stringent test of the flux measurement method.

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Fluxes are computed from ten-minute data segments of CO and vertical wind velocity data with 50 % overlap of each segment (i.e. 11 ten-minute segments per hour). The linear trend is subtracted from each segment and a Hamming window is applied to limit leakage of low frequency variance unrelated to turbulent flux. The slope from the linear trend in CO is retained as a measure of $\partial \mathrm{CO}/\partial t$ for each segment. Flux is computed as the area of the cospectrum. Based on an analysis of DMS flux cospectra at similar frequency response and wind speed (Yang et al., 2011), a 4 % adjustment is applied for unsampled flux signal above 2 Hz.

Ten-minute flux results are selected to eliminate periods unsuitable for eddy correlation measurements. In this case relevant selection criteria are relative wind direction within ±60°, standard deviation in relative wind direction < 10°, and heading change less than 25° in ten minutes. In addition, it was necessary to impose criteria limiting the magnitude of $\partial CO/\partial t$ and horizontal turbulent flux, as described in Sect. 4.

Figure 3 shows flux results for the two days on station, bin averaged to hour-of-day (local time). The diel CO flux cycle in Fig. 3 closely mirrors the cycle in seawater concentration in Fig. 2, with a pre-dawn minimum and early afternoon maximum.

For a mean seawater CO concentration of 1.5 nM at the peak in the flux diel cycle (2–3 PM local time), the computed CO transfer coefficient (k_{660}) is 41 $\mathrm{cm\,h^{-1}}$ at $U_{10} \sim 11\,\mathrm{m\,s^{-1}}$. For these conditions the Wanninkhof (1992) model gives $k_{660} = 38\,\mathrm{cm\,h^{-1}}$; Nightingale et al. (2000) is $k_{660} = 29\,\mathrm{cm\,h^{-1}}$; McGillis et al. (2001) is $k_{660} = 40\,\mathrm{cm\,h^{-1}}$; and Woolf (1997) yields $k_{660} = 40\,\mathrm{cm\,h^{-1}}$ (solubility, Schmidt number and transfer velocities computed using R-scripts of Johnson, 2010). Clearly, with limited data it is impossible to draw too much from this agreement, but the flux magnitude is close to the expected value. Improvements in frequency response and a careful analysis of signal attenuation would further limit errors from lost signal above 2 Hz. More extensive sampling of the seawater CO concentration is necessary to critically assess the transfer coefficient.

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4 Sources of flux uncertainty

Rowe et al. (2011) have analyzed sensor resolution requirements for eddy correlation surface flux measurements. For the conditions of this study and the noise level of the CO analyzer, their analysis predicts a 60 % random error for hourly average CO flux measurements at a seawater concentration of 0.5 nM. In fact, scatter in hourly CO flux from midnight to 06:00 a.m. local time (when seawater CO is ∼0.5 nM) suggests an error three times greater: $0.00063 \pm 0.00118\,\mathrm{ppb\,m\,s^{-1}}$, or 187 % relative standard deviation.

The expected scalar variance from surface flux alone may be predicted from similarity theory as a function of friction velocity, u_* (Blomquist et al., 2010; Fairall et al., 2000). Assuming neutral stability, the relationship takes the form $\sigma_c = 3|\overline{w'c'}|/u_*$. Table 1 compares observed and similarity-predicted variance for DMS and CO flux measurements. Observed DMS variance from a cruise in the Sargasso Sea is quite close to the similarity estimate. This is reasonable as the sea surface is the sole source of DMS and its atmospheric lifetime is sufficiently short (2–3 days) to limit the influence of distant sources. Results for CO show an observed variance (minus sensor noise) four times greater than the similarity-predicted surface flux variance.

Even under clean background conditions in the remote marine boundary layer, where the relative standard deviation in ten-minute mean CO concentration was just 2 % over two days on-station, small gradients in CO concentration may yield significant variance from horizontal turbulent flux many times greater than from surface flux. Figure 4 illustrates the relationship between components of the horizontal turbulent flux and $\partial \mathrm{CO}/\partial t$. A positive correlation exists between the along-wind component of horizontal flux ($\overline{u'c'}$) and $\partial \mathrm{CO}/\partial t$, indicating advection of the CO gradient past the ship. Many samples have horizontal fluxes quite large compared to the magnitude of surface flux in Fig. 3. For this reason, additional criteria were applied to eliminate ten-minute segments with excessive gradient influence: specifically $|\partial \mathrm{CO}/\partial t| < 2.7\,\mathrm{ppb\,h^{-1}}$ and

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$|\overline{u'c'}| < 0.026\,\mathrm{ppb\,m\,s^{-1}}$ (the 80 % confidence limit of the mean in each case, shown as a bounding box in Fig. 4).

Figure 5 presents cospectra for $\overline{w'c'}$ and $\overline{u'c'}$, representing mean fluxes for selected early afternoon ten-minute segments (35 segments, 01:00–04:00 p.m. local time). Even removing outliers, $\overline{w'c'}$ cospectra are noisy due to residual variance contributed by horizontal turbulent flux which is several times greater than vertical flux. The large component of horizontal flux at low frequencies is mirrored in the CO variance spectrum (Fig. 1). The mean absolute horizontal flux for ten-minute segments meeting selection criteria (mean $|\overline{u'c'}| = 0.008\,\mathrm{ppb\,m\,s^{-1}}$ and mean $\overline{u'c'} = -0.0034 \pm 0.0091\,\mathrm{ppb\,m\,s^{-1}}$) is eight times greater than the mean vertical flux of $0.001\,\mathrm{ppb\,m\,s^{-1}}$.

The variance budget production term associated with scalar flux in a mean gradient is (Stull, 1988)

$$-2\overline{u_i'c'}\frac{\partial \overline{C}}{\partial x_i} \tag{1}$$

where u_i specifies the full turbulent wind field. From similarity theory, an estimate for the vertical scalar gradient is

$$\frac{\partial \overline{C}}{\partial z} = -\frac{\overline{w'c'}}{\kappa u_* z} \tag{2}$$

which is $\sim 3.5\times10^{-4}\,\mathrm{ppb\,m^{-1}}$ ($0.35\,\mathrm{ppb\,km^{-1}}$) for mean conditions of this test (mean flux $= 0.001\,\mathrm{ppb\,m\,s^{-1}}$, $u_* = 0.4\,\mathrm{m\,s^{-1}}$). Thus, term (1) for horizontal flux becomes comparable to the equivalent term for vertical flux when $\partial C/\partial x \sim 4.3\times10^{-5}\,\mathrm{ppb\,m^{-1}}$ (4.3 ppb per 100 km), which is a low threshold gradient for a species with a mean background concentration of 60–150 ppb. At $\overline{U} = 10\,\mathrm{m\,s^{-1}}$ this corresponds to $|\partial \mathrm{CO}/\partial t|$=1.5 $\mathrm{ppb\,h^{-1}}$ (half the 2.7 $\mathrm{ppb\,h^{-1}}$ selection criterion). Selection criteria therefore limit scalar variance from horizontal turbulent flux to approximately twice the variance from surface flux. In this test, these limits seem sufficient to eliminate the majority of outlier measurements.

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The observation that additional sources of variance degrade precision of the CO surface flux measurement imposes stringent location selection criteria for future CO air-sea flux studies. Conditions at station ALOHA appear suitable, and on that basis many locations in the Southern Hemisphere should also be acceptable.

5 Conclusions

The analytical performance of a commercially available infrared OA-ICOS trace gas analyzer is sufficient for ship-based flux measurements of CO at moderate to high wind speeds when seawater concentration is >1 nM. A clear diel cycle in CO flux, mirroring the cycle in seawater concentration, was observed over two days at a research site near Oahu in the oligotrophic North Pacific subtropical gyre. CO flux measurements by eddy correlation are a potentially important development for investigations of biogeochemical and physical dynamics in the ocean's surface mixed layer. Additionally, CO is an important low solubility end-member in the spectrum of gases involved in air-sea exchange. As such, it should exhibit significant bubble-mediated gas exchange enhancement at moderate-to-high wind speeds, providing an interesting and important test of physical gas transfer theory.

The moderately long CO atmospheric lifetime (~ 50 days), combined with vigorous natural and anthropogenic sources, yields a high, variable background atmospheric concentration of ~ 100 ppb in the N Hemisphere and half that value in the S Hemisphere. CO variance from horizontal turbulent diffusion of atmospheric gradients as small as 1–2 % of the mean concentration per 100 km reduces precision of the eddy correlation measurement. This places a premium on selecting study sites in remote marine locations with minimal variability in the background concentration.

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Table 1. Comparison of parameters for DMS and CO flux measurement.

Parameter	DMS	CO	Notes
Mean atm. conc. (ppb)	0.100	100	
Atm. lifetime (d)	2–3	50	
Sea-air ΔP (ppb)	6.6	1668	[DMS] = 2.6 nM, [CO] = 1.5 nM
SST (°C)	28	22	
Solubility, α	8.9	0.019	dimensionless liq/gas
U_{10} ($m\,s^{-1}$)	6	11	
k ($cm\,h^{-1}$)	10	45	at ambient SST and Sal
u_* ($m\,s^{-1}$)	0.2	0.4	
Flux ($ppb\,m\,s^{-1}$)	0.0016	0.0040	$F = \alpha k \Delta P$
σ_{sim} (ppb)	0.021	0.030	from similarity
σ_{obs} (ppb)	0.020	0.126	observed minus sensor noise
$\sigma_{sim}/\sigma_{obs}$	1	0.24	

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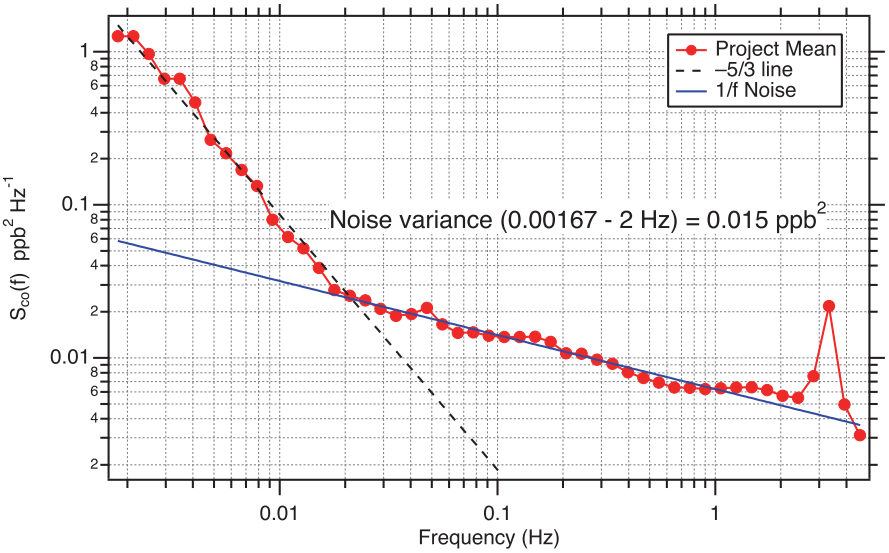

Fig. 1. Mean bin averaged CO variance spectrum for 10-min data segments at sea. Valid data segments were selected using criteria described in Sect. 3. The peak at 3 Hz is an analyzer artifact.

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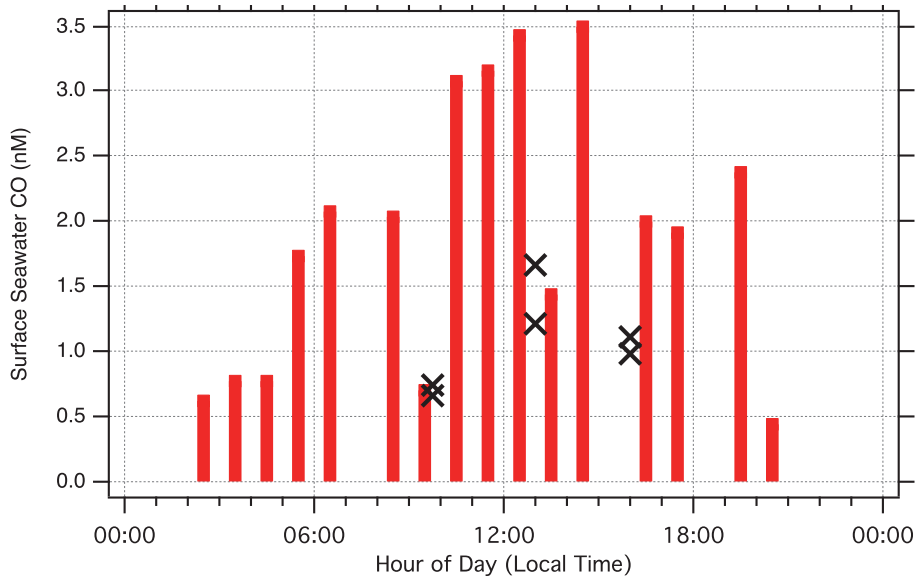

Fig. 2. Diel cycle in seawater CO at station ALOHA (local time). Crosses: this cruise. Bars: bin averaged results from previous HOT cruises over all seasons since 2008.

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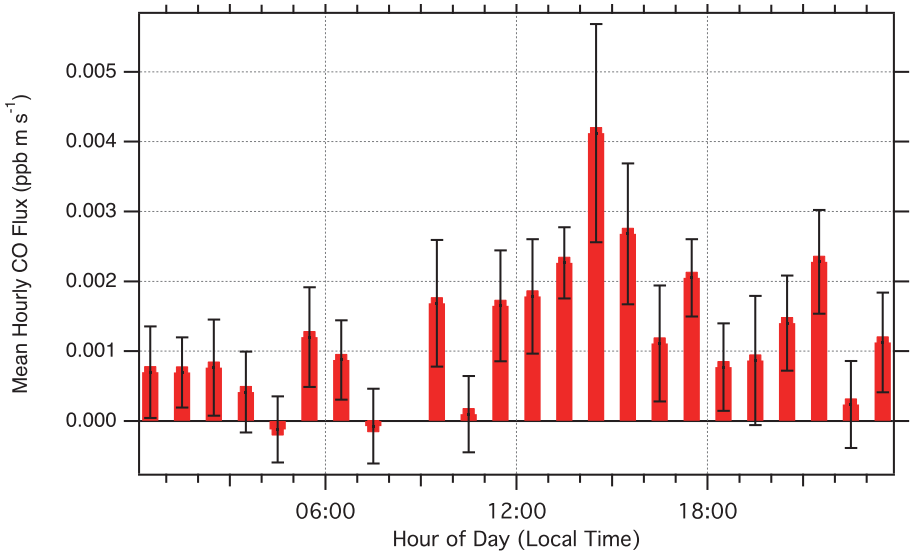

Fig. 3. Diel cycle in CO flux over a two day period at station ALOHA. Error bars are standard error of the mean.

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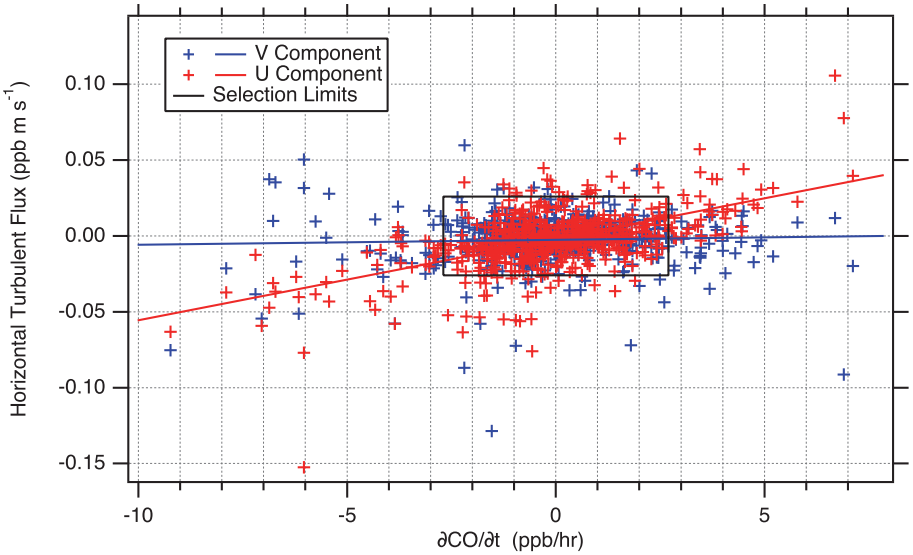

Fig. 4. Trend in $\partial \mathrm{CO}/\partial t$ and along-wind ($\overline{u'co'}$) and cross-wind ($\overline{v'co'}$) components of horizontal turbulent flux. The bounding box gives the 80 % confidence limit of the mean in each variable. Results outside the bounding box are excluded on the basis of selection criteria, limiting the excessive influence of CO variance from non-surface-flux sources.

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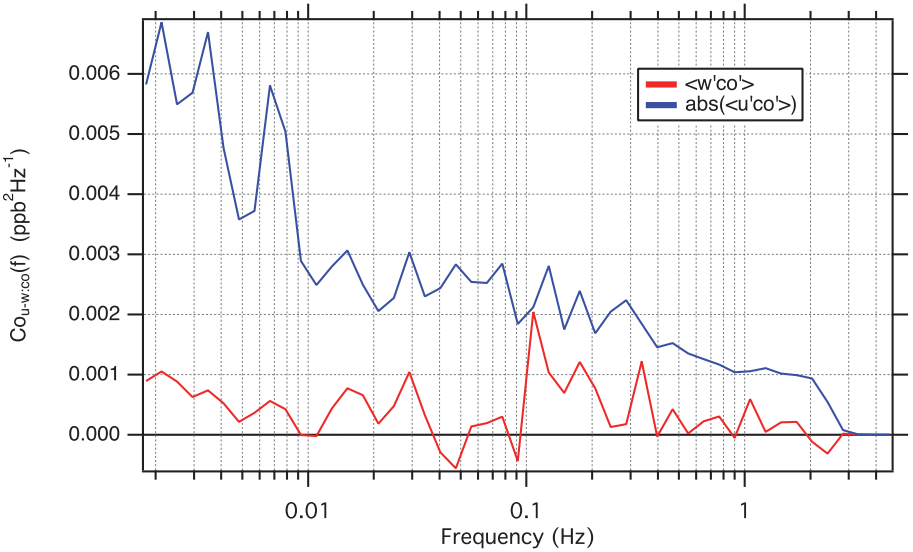

Fig. 5. Mean cospectra illustrating vertical ($\overline{w'co'}$, red) and horizontal ($|\overline{u'co'}|$, blue) CO turbulent flux for selected ten-minute afternoon segments (01:00–04:00 p.m. local time). Horizontal flux is computed as the mean of absolute values.

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