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Atmospheric aerosol light scattering and surface wetness influence the diurnal pattern of net ecosystem exchange in a semi-arid ponderosa pine plantation

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Abstract

The diurnal variation of net ecosystem exchange (NEE) showed an unusual pattern at the Blodgett Forest Ameriflux site, with late afternoon NEE lower than early morning (indicating more uptake), while air temperature and atmospheric vapor pressure deficit were much higher. To investigate processes influencing this pattern, NEE was compared to several environmental variables during summer 2002. Unusual variations of NEE can be partly attributed to dew formation on the leaf surface. An empirical model is used to show that surface wetness reduced the net ecosystem uptake of CO_2 during the morning by 11%. In addition, transport of air-pollution from the Central Valley to this site results in higher aerosol particle concentration, light extinction and light scattering during the afternoon than in the morning. Total irradiance was 11% lower during the afternoon than in the morning, while diffuse irradiance was 24% higher. The empirical model is used to show that the decrease in total radiation reduced photosynthesis during the afternoon, but the increase in diffuse radiation enhanced photosynthesis even more. Aerosol loading caused net uptake of CO_2 by the forest to increase by 8% in the afternoon as a result of changes in direct and diffuse radiation.

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1. Introduction

An unusual diurnal pattern in net ecosystem exchange of CO_2 is consistently observed at the

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Blodgett Forest Ameriflux site. Late afternoon net CO_2 uptake by the vegetation is higher than early morning, in contrast to what would be expected based on the diurnal variation of variables normally considered to control CO_2 flux. Previous studies have shown that photosynthetically active radiation, temperature, and vapor pressure deficit are the main variables controlling the diurnal pattern of net

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ecosystem exchange (e.g. Baldocchi, 1997). Relationships using these variables have been used to model carbon exchange between the atmosphere and vegetation, and to quantify terrestrial carbon sequestration (Bonan, 1995). Others studies suggest that additional factors such as aerosol loading and dew formation on the leaf surface could also play an important role (Niyogi et al., 2004; Hanba et al., 2004).

Under diffuse light, radiation inside the canopy reaches leaves more evenly and fewer leaves are lightsaturated, which increases light use efficiency of canopy photosynthesis (Hammer and Wright, 1994; Oechel and Lawrence, 1985; Sinclair et al., 1992). Clouds and natural aerosols such as volcanic dust affect the proportion of diffuse radiation and can enhance terrestrial carbon sequestration (Gu et al., 1999, 2002, 2003; Roderick et al., 2001). Anthropogenic aerosols have more efficient optical extinction for light than natural aerosols (Carrico et al., 2003; Seinfeld and Pandis, 1998) and are responsible for about half of the extinction and scattering of light by particles globally (IPCC, 2001). Recently, Niyogi et al. (2004) reported the first observational evidence of a link between routine aerosol variability, diffuse radiation and carbon sequestration by different types of vegetation.

Nighttime temperature inversions are common in Mediterranean climates. Inversions develop during stable conditions, as heat loss by radiation causes the land surface temperature to fall quickly (McIntosh and Thom, 1983). When the air in contact with the ground reaches its dew point temperature, water vapor begins to deposit on surfaces as dew. In some plants, stomata close completely after artificially increasing leaf surface wetness by misty rain (Ishibashi and Terashima, 1995), which would reduce uptake of CO₂. Dew on the leaf surface could also reduce the diffusion of CO_2 from the atmosphere to the sub-stomatal cavity (Brewer and Smith, 1995). The extent to which these mechanisms affect ecosystem scale CO₂ uptake by coniferous forests has not been reported in the literature.

In this paper, we present a case study investigating the effects of anthropogenic aerosols and dew formation on the diurnal pattern of net ecosystem exchange. Both processes are potentially important at the Blodgett Forest because of its location in a semiarid mountainous climate on the western slope of the Sierra Nevada Mountains of California. This site is situated downwind of significant anthropogenic pollution sources in Sacramento and the agricultural Central Valley of California, which is shown to significantly affect the amount of direct and diffuse radiation.

2. Materials and methods

The site and most of the measurements have been described in Goldstein et al. (2000) and Xu et al. (2001) and will be summarized focusing on the important characteristics relevant to this paper. Data presented here were obtained during the summer and fall 2002 (May to October). A list of symbols and acronyms is presented in Appendix A.

2.1. Site description

The Blodgett Forest Ameriflux site was established in a young ponderosa pine plantation on the western slope of the Sierra Nevada Mountains of California in 1997. It is located ($38^{\circ}53'42.9''$ N, $120^{\circ}37'57.9''$ W) at 1315 m above sea level, on land owned by Sierra Pacific Industries. Trees were planted in 1990 at a density of ~1200 trees per hectare. In 2002, the plantation consisted of 12-year-old trees with a ~4.4 m average canopy height dominated by ponderosa pine. The major understory shrubs were *Arctostaphylos* spp. and *Ceonothus* spp. The leaf area index (LAI) of ponderosa pine and the shrubs in summer 2002 was ~2.2 and ~0.9 m² m⁻², respectively.

The site is characterized by a Mediterranean climate, with warm dry summers and cold wet winters. In 2002, the daily mean air temperatures ranged from -5 to 20 °C in winter and from 14 to 30 °C in summer. The daily mean vapor pressure deficit (VPD) ranged from 0.0 to 1.5 kPa in winter and from 1.0 to 3.5 kPa in summer. In 2002, no precipitation occurred from May 21st to November 6th, and soil water content at 10 cm deep decreased from 35.8% in mid-May to 9.7% in early November. Total precipitation in 2002 was 1233 mm (696 mm before mid-May).

The daytime airmass trajectory at the site is very consistent and comes from the South-West, directly upslope from the Sacramento region. This regular transport pattern brings anthropogenic air-pollution to the site in the afternoon (Dillon et al., 2002; Lamanna and Goldstein, 1999). At night, air flows downslope from the Sierra Nevada Mountains (North-East), flushing the site with cleaner air.

2.2. Canopy scale measurements

In 2002, wind velocity and virtual temperature fluctuations were measured at 10 Hz with a threedimensional sonic anemometer (ATI Electronics Inc., Boulder, CO) mounted on a horizontal beam on a tower at 10.5 m above the ground. CO_2 and H_2O mixing ratios were measured with an infrared gas analyzer (Li-Cor 6262, Lincoln, NE). Ambient air was sampled 10 cm downwind from the sonic anemometer at $5.5 \,\mathrm{l\,min}^{-1}$ through 18 m of 4 mm inside diameter Teflon tubing. The sampling tube was not heated. Additionally, the canopy vertical profile of CO₂ and H₂O mixing ratios were measured every 30 min by sampling sequentially at five heights for 6 min each. Half-hour fluxes of CO₂, H₂O, and sensible heat were determined by the eddy covariance method (Baldocchi et al., 1988; Shuttleworth et al., 1984). Fluxes were calculated using ARAMF 2.0 (Advanced Routines for the Analysis of Measurements Flights, ARA, Australia). Negative fluxes indicate mass and energy transfer from the atmosphere to the surface.

The sonic anemometer data was rotated to force the mean vertical wind speed to zero, and to align the horizontal wind speed onto a single horizontal axis. The calculated vertical rotation angle was typically 0.6° and did not vary by more than a few degrees. The time lag for sampling and instrument response was determined by maximizing the covariance between vertical wind velocity (w') and scalar (c') fluctuation. Typical values were 3.3 s in 2002 and were extremely consistent throughout the measurement period. Errors due to sensor separation and damping of high frequency eddies were corrected using spectral analysis techniques as outlined by Rissmann and Tetzlaff (1994). Spectral analysis revealed an underestimation of gas fluxes of roughly 9% for CO₂ and 12% for H₂O. Correction factors for each half-hour were calculated for each gas and applied to the fluxes only during the times when the sensible heat flux data were reliable (97% of the data). In some studies of CO_2 fluxes over canopies, a short burst of positive CO_2 flux can be seen in the early morning due to the 'flush out' of CO_2 as the nighttime inversion layer breaks down (Grace et al., 1995). Our site exhibited no such 'flush out'; furthermore, measurements of intracanopy CO_2 storage confirm that nocturnal CO_2 storage was small in this ecosystem (Goldstein et al., 2000). However, storage corrections were applied to the fluxes of CO_2 in order to avoid underestimation during any critical period of the day (Aubinet et al., 2000). Two separate model estimates of the flux footprint agreed that 90% of the daytime footprint was within 200 m of the tower (Baker et al., 1999).

2.3. Aerosol measurements

During summer 2002 (May 9th-November 14th) aerosols were measured using a variety of instruments. The optical properties of the aerosols were measured with an aethelometer (model AE2, McGee Scientific, Berkeley, CA) and an integrating nephelometer (NGN-2, OPTEC Inc., Lowell, MI). An integrated measure of the number concentration of aerosol particles was provided by a condensation particle counter (CPC TSI 3022A, TSI Inc., Shoreview, MN). Particle sizes were measured using an optical particle counter (Lasair 1003, Particle Measurement Systems, Boulder, CA) providing size distributions from 0.1 to 10 µm, and a scanning electrical mobility spectrometer (SEMS) providing size distributions between 10 and 400 nm. The SEMS system utilized a differential mobility analyzer (TSI 3071A) coupled with a CPC (TSI 3760) as a detector. All instruments except the Lasair 1003 were mounted on the tower, in individual weatherproof containers. The Lasair 1003 was located in the adjacent temperature controlled instrument building. The air was sampled at 16.7 l min⁻¹ through a PM10 inlet followed by a PM2.5 sharp cut cyclone (models 57-000596 and 57-005896, respectively, Rupprecht & Patashnick Co., Albany, NY). The inlet was mounted above the top of the tower at a height of 12 m. Sample flows were isokinetically sampled from the main flow.

The nephelometer measured the light scattering coefficient at an effective wavelength of 550 nm. The instrument, described in more detail by Molenar et al. (1989) had an open-air design, allowing the air to pass through an open door in the side of the instrument. This design minimized the changes in relative humidity and temperature of the air as it enters the scattering chamber, a phenomenon that has lead to underestimated scattering measurements in other nephelometers. The instrument allowed a wide spectrum of particle sizes, including particles greater than 2.5 mm, to pass into the measurement chamber due to its open design. However, the maximum particle detectable by the instrument has not been characterized. The aethelometer measured light absorption of the aerosol using a filter-based light attenuation technique (Hansen et al., 1984). The instrument used at our site measured light absorption at two wavelengths, 880 nm (near IR) and 350 nm (near UV). Light extinction is a function of both light scattering and absorption in the atmosphere. However, diffuse radiation is only a function of light scattering. Field experiments showed that aerosols could be present in distinct layers at different heights (Seinfeld and Pandis, 1998). The measured light scattering coefficient only took into account light scattered by aerosols present at the top of the canopy.

2.4. Environmental measurements

In 2002, environmental parameters were recorded every 5 s and half-hour averages were calculated and stored on a datalogger (see Goldstein et al. (2000) for description of all instruments). The parameters included wind direction and speed, vertical profile of wind speed at three heights, vertical profile of air temperature and humidity, net radiation, photosynthetically active radiation, soil temperature in three locations at 5, 10, 15, 30 and 50 cm depth, soil moisture at 10, 30 and 50 cm depths, soil heat flux at three locations, rain, and atmospheric pressure.

The surface wetness was recorded by four Model 237 surface wetness sensors (Campbell Scientific Inc., Logan, UT) installed on branches at ~2.0 m height. The Model 237 is an artificial electrical resistance sensor. Condensation on the sensor lowers the resistance between copper fingers on circuit board mounted on a flat plate. The sensor resistance varies from above 3,000,000 Ω when totally dry to around 1000 Ω when totally wet. Based on the resistance measured, surface wetness was normalized on a scale where 0% represents totally dry and 100% represents

totally wet. Flat horizontal plates can collect dew differently to needles, so the sensors were exposed vertically. Leaf biomass measurement showed that height of the sensors (2.0 m) was about the mid-height of the canopy (4.5 m), and where most of the leaf surface was located (Xu, 2000).

Diffuse photosynthetically active radiation (I_f) was not measured during summer 2002 concurrently with the aerosol properties. Measurement of I_f (BF3, Delta-T, Cambridge, UK) started in summer 2003. Following the approach of Gu et al. (2002), we used a radiation-partitioning model to calculate diffuse and direct PAR during summer 2002. This model was parameterized using data collected in 2003. Results are shown and discussed in Appendix B. In contrast with the nephelometer, which measured light scattering at the top of the tower only, the BF3 sensor measured diffuse PAR as a result of scattering in the entire atmospheric column between the land surface and the sun.

3. Results and discussion

NEE, meteorological and aerosols data simultaneously covered the period from May 9th to November 14th 2002. However, data presented in this paper has been limited to May 21st through October 23rd 2002. This period was chosen because: (1) it was free of precipitation which makes all the measurements more reliable, (2) photosynthetically active radiation (PAR), air temperature and vapor pressure deficit (VPD) did not show abrupt variations due to seasonal changes.

3.1. Clear-sky and cloudy conditions

Passing weather fronts and cloudy conditions greatly influence meteorological variables such as air and soil temperature, PAR, VPD and the proportion of diffuse versus direct radiation. All these variables affect NEE. Therefore, cloudy conditions could potentially confound the effects of aerosol loading and surface wetness. In order to focus on these effects, clear-sky days were segregated from cloudy days in the available dataset. Following the methodology of Gu et al. (1999), (99) days with smoothly changing values of PAR and clearness index (k_t , see Eq. (B.1)) Table 1

Mean diurnal values of net CO₂ flux and meteorological variables for clear-sky and cloudy conditions during summer 2002 (21st May–23rd October)

	NEE (μ mol m ⁻² s ⁻¹)	PAR (μ mol m ⁻² s ⁻¹)	$I_{\rm f}/I_{\rm t}$ (-)	$T_{\rm a}$ (°C)	VPD (kPa)	W _s (%)
Clear-sky $(n = 1894)$	-6.7	1236	0.25	22.2	1.9	5.7
	(0.1)	(12)	(0.003)	(0.1)	(0.01)	(0.2)
Cloudy $(n = 881)$	-7.2	1152	0.35	19.2	1.4	6.9
	(0.1)	(19)	(0.007)	(0.2)	(0.02)	(0.3)

NEE: net ecosystem exchange, PAR: photosynthetically active radiation, I_t/I_t : fraction of diffuse radiation, T_a : air temperature, VPD: vapor pressure deficit, W_{s} : surface wetness. Standard errors of the mean are shown in parenthesis (clear-sky days n = 99, cloudy days n = 49).

were selected as clear-sky days, and 49 days were classified as cloudy (66 and 33%, respectively).

Diurnal patterns of NEE, meteorological variables and aerosol properties (concentration and scattering) were similar for cloudy and clear-sky days (not shown), but the mean values differed (Table 1). PAR, air temperature and VPD were lower on cloudy days, while the fraction of diffuse radiation and surface wetness was higher. The net uptake of CO_2 by the forest was 0.5 μ mol m⁻² s⁻¹ higher on cloudy days. Cloudy conditions only affected mean values but did not affect the diurnal patterns of NEE, meteorological variables and aerosol properties, which are the focus of this paper. Therefore, all days were pooled together for the subsequent analysis in order to increase the number of available data.

3.2. Diurnal pattern of NEE

We plotted the mean diurnal pattern of NEE as a function of PAR to better identify the controlling processes (Fig. 1). During early morning, NEE was near zero; then later in the morning NEE became negative indicating net carbon uptake by the vegetation (Fig. 1a). In the early afternoon (<16:00 h PST) there was less carbon uptake than in the morning for the same incoming PAR. Then in the late afternoon (>16:00) there was more carbon uptake than in the early morning for the same incoming PAR.

NEE is the difference between ecosystem respiration (ER) and gross ecosystem productivity (GEP) (or canopy photosynthesis):

$$F_{\rm NEE} = F_{\rm ER} - F_{\rm GEP} \tag{1}$$

where F_{NEE} is the net ecosystem exchange carbon flux, F_{ER} is ecosystem respiration and F_{GEP} is gross ecosystem productivity. Typically NEE shows increasing carbon uptake with increasing PAR because canopy photosynthesis responds positively to incoming radiation. However, there is usually less net uptake of carbon in the afternoon than in the morning (Anthoni et al., 1999, 2002; Arneth et al., 1998; Baldocchi, 1997; Baldocchi et al., 1997; Falge et al., 2002; Granier et al., 2002; Hunt et al., 2002; Schmid et al., 2000). This is partly due to warmer air and soil temperature in the afternoon promoting ecosystem respiration (Anthoni et al., 1999; Bowling et al., 2001, 2003; Xu et al., 2001). Furthermore, many studies have shown that higher VPD in the afternoon promotes partial stomatal closure decreasing canopy photosynthesis, especially in ecosystems where low soil moisture cannot support optimal rates of carbon assimilation throughout the day (Anthoni et al., 1999, 2002; Arneth et al., 1998; Baldocchi, 1997; Baldocchi et al., 1997; Falge et al., 2002; Granier et al., 2002; Hunt et al., 2002; Schmid et al., 2000).

In the same way, VPD was consistently higher in the afternoon than in the morning at our site (Fig. 1d), which would decrease stomatal conductance and impose constraints on GEP. In addition air and soil temperature were much higher in the afternoon than in the morning, which would enhance ER in the afternoon (Fig. 1b and c). Xu et al. (2001) and Tang et al. (2004) fitted different models of soil and ecosystem respiration at our site based on chamber data. They found the response of ER to soil temperature followed an exponential relationship with a Q_{10} = 1.6, which supports our assumption that ER is higher in the afternoon than in the morning. The above mentioned effects of VPD and temperature on NEE explain why our data show that the net uptake of CO_2 at our site was less in the early afternoon (<16:00) than in the late morning (>08:00) for the same incoming radiation (Fig. 1a). However, these effects



Fig. 1. Mean diurnal pattern of (a) NEE, (b) air temperature, (c) soil temperature at 5 cm, (d) VPD, (e) fraction of diffuse PAR, (f) surface wetness as a function of PAR during the summer 2002. Standard errors of the mean and hours of day are shown.

are inconsistent with our data showing that the net uptake of CO_2 was larger in the late afternoon (>16:00) than in the early morning (<08:00) for the same incoming radiation (Fig. 1a). This variation is unexpected and has not been previously reported in the literature.

Experimental artifacts could explain this unusual pattern, such as systematic differences in the footprint or turbulence statistics between the morning and the afternoon. Small variation in the footprint existed because the main wind direction was slightly different (morning $\cong 180^\circ$, afternoon $\cong 250^\circ$). However, the vegetation was not significantly different in the two directions. In addition, the unusual variation of NEE was still evident when selecting data only from the mean daytime direction $(210 \pm 20^{\circ})$. Systematic variations in turbulence parameters such as the friction velocity (u^*) existed between the morning $(u^* \cong$ 0.35 m s⁻¹) and the afternoon $(u^* \cong 0.44 \text{ m s}^{-1})$. Nevertheless, most of the daytime u^* values were greater than 0.2 m s^{-1} , which characterize good mixing conditions that validate the use of the eddy covariance method in this open canopy ecosystem. Furthermore, the diurnal pattern of NEE was unaffected by selecting only data with $u^* > 0.2 \text{ m s}^{-1}$. Other factors must have affected NEE during the morning (suppression of net uptake) and/or during the afternoon (enhancement of uptake) to cause the observed pattern. In this paper, we investigate how diffuse radiation and surface wetness relate to the observed daily variation in NEE.

3.3. Diurnal pattern of surface wetness

Fig. 1f shows that surface wetness as measured by the wetness sensors was much higher during the early morning (<08:00) than during the late afternoon. Fig. 2 shows the relationship between surface wetness and NEE at low PAR (<500 μ mol m⁻²s⁻¹), with data sorted by early morning and late afternoon periods. During both periods, the net uptake of CO₂ by the vegetation was suppressed when surface wetness was greater than 10%. When surface wetness was below 10%, larger variations of NEE existed and were most likely controlled by variations of other meteorological



Fig. 2. Relationship between surface wetness and NEE. Data were selected for low par level (<500 μ mol m⁻² s⁻¹). Open symbols are for morning values and closed symbols for afternoon values.

variables. Furthermore surface wetness was higher during the morning with values often exceeding 40%. Fig. 3a compares the mean diurnal patterns of NEE based on the complete dataset (as in Fig. 1a) with the pattern based on a sub-dataset only including values when surface wetness was below 5%. Most of the diurnal pattern is similar, apart from variations during the early morning where NEE showed more net uptake for data with low surface wetness. These results suggest that dew in the morning suppressed net uptake of CO_2 by this ecosystem.

Surface wetness impacts on leaf photosynthesis vary by species and habitats (Brewer and Smith, 1997; Brewer et al., 1991; Smith and McClean, 1989). Wettability is an important factor. Enhanced photosynthetic response to wetness occurs in non-wettable leaf species and is due to changes in stomatal regulation (Hanba et al., 2004). Water droplets may reduce the vapor pressure deficit at the leaf surface, increasing leaf water potential and stimulating stomatal conductance and photosynthesis (Hanba et al., 2004; Smith and McClean, 1989). However, suppressed photosynthetic response to wetness occurs on leaves of wettable species resulting from reduction of stomatal conductance (Hanba et al., 2004; Ishibashi and Terashima, 1995) and/or decrease in diffusion, since CO₂ diffusivity in water is 1000 times lower than in air (Brewer and Smith, 1995; Grantz et al., 1997; Kato et al., 2002). For example, laboratory experiments on Phaseolus vulgaris L. have shown that stomata closed completely after artificially increasing leaf surface wetness by misty rain (Ishibashi and Terashima, 1995). Stomata gradually opened to half the original aperture after 60 min. The rate of CO_2 exchange measured on the wet leaves changed in concert with the changes in stomatal aperture, attaining 60% of the control level after 1 h (Ishibashi and Terashima, 1995). In the case of long term exposure to dew on a wettable species, biochemical



Fig. 3. Mean diurnal pattern of NEE. Open symbols: all data (n = 2896, as in Fig. 1a). (a) Closed symbols: data with leaf wetness <5% (n = 2355), (b) closed symbols: data with vapor pressure deficit <1.5 kPa (n = 1098).

inhibition has been shown to occur (Hanba et al., 2004; Ishibashi et al., 1997). In general, large and flat leaves are considered wettable, while smooth and waxy leaf surface such as ponderosa pine needles are considered non-wettable. However, our field experience of leaf gas exchange chamber measurements at Blodgett tell us that on most summer mornings ponderosa pine needles are covered with water that stay on the surface until radiational heat exchange is strong enough to evaporate it. A redefinition of the "wettable" character of ponderosa pine needles might be necessary.

3.4. Aerosol light extinction and scattering

Particle number density exhibited a regular diurnal pattern with a minimum at approximately 05:00 h PST, increasing after 10:00, reaching a plateau around 17:00, and decreasing during the evening (Fig. 4a). Light scattering showed the same diurnal pattern, with stable values during the morning and an increasing trend during the afternoon (Fig. 4b). The clearness index was stable during the morning and decreased during the afternoon (Fig. 4c). PAR was systematically lower during the afternoon than in the morning for the same solar elevation angle, with a maximum difference of 187 μ mol m⁻² s⁻¹ (84 W m⁻²) at a solar elevation angle of 30° (Fig. 4d). Average total PAR in the afternoon was 11% lower than in the morning.

Fig. 1e shows higher diffuse PAR fraction in the afternoon than in the morning for the same total incoming radiation. The diffuse PAR fraction was 0.36 (174 μ mol m⁻² s⁻¹) and 0.54 (267 μ mol m⁻² s⁻¹) at 06:30 and 17:00, respectively, while total PAR was about 490 μ mol m⁻² s⁻¹. Average diffuse PAR in the afternoon was 24% higher than in the morning.

In other studies, the clearness index was correlated with the solar elevation angle, because the path length through the atmosphere is one of the factors affecting direct radiation absorption (Ineichen and Perez, 1999; Perez et al., 1990). In addition, Gu et al. (1999) reported that, in two temperate ecosystems, the afternoon clearness index was generally larger than the morning for a given solar elevation angle. Our data show a different pattern, with a stable clearness index during the morning for increasing solar elevation angles, and a decreasing clearness index during the afternoon while solar angle decreases. Consequently the clearness index was larger in the morning than in the afternoon, in contrast with data presented by Gu et al. (1999).

The diurnal pattern of total and diffuse PAR fraction was correlated with the variations of aerosol concentration, light scattering and clearness index. These results reflect the particular pattern of airpollution at our site. Blodgett Forest has a sky with very clear background conditions due to it midrange Sierra Nevada mountain elevation (1375 m) and Mediterranean climate. Transport of air-pollution is controlled by the extremely consistent diurnal pattern of winds that carry polluted air upslope from the Central Valley into the Sierra Nevada by day, and flush the mountains downslope at night with cleaner continental background air (Dillon et al., 2002; Lamanna and Goldstein, 1999). Movement of urban aerosols transported to the mountains by day explains much of the diurnal pattern observed in the particle concentration, total and diffuse PAR at our site. Furthermore, previous research has shown that the reaction of O₃ with biogenically-emitted hydrocarbons is prevalent in this area and likely leads to secondary aerosol growth (Goldstein et al., 2004; Holzinger et al., 2004; Kurpius and Goldstein, 2003). This process may also influence the diurnal pattern of particle concentration, total and diffuse PAR, by contributing an additional biogenic component to the urban aerosol in the afternoon.

Stanhill and Cohen (2001) estimated that irradiance has declined globally by 11% in average over the past 50 years due to increased in cloudiness and aerosol concentration. We report that, on average, total PAR in the afternoon was 11% lower than in the morning due to the increase in aerosol number density at our site. Aerosol light extinction of this magnitude is likely to influence the carbon balance of the vegetation. The expected change from reduced total PAR would be a decrease in canopy photosynthesis. On the other hand, several studies have shown that diffuse light, which enters a complex canopy from all directions, reaches leaves more evenly than light in the direct solar beam (Hammer and Wright, 1994; Sinclair et al., 1992). Consequently, canopy light use efficiency increases with fraction of diffuse radiation and canopy photosynthesis has less tendency to saturate (Gu et al., 2002). Differences between early morning and late afternoon



Fig. 4. Mean diurnal pattern of (a) particle count, (b) light scattering, (c) clearness index, and (d) PAR as a function of the solar elevation angle during the summer 2002 (22nd May–22nd October). Standard errors of the mean and hours of day are shown.

NEE at our site could be partly due to enhanced photosynthesis caused by the increase in the fraction of diffuse radiation in the afternoon. Recently, Niyogi et al. (2004) presented the first direct, multisite observations showing that relatively routine aerosol variability affects the regional terrestrial carbon cycle. In particular their results suggest that the CO_2 sink increases with aerosol loading for forest and crop lands and decreases for grassland (Niyogi et al., 2004).

3.5. Empirical model of NEE

In order to quantitatively test the hypotheses that aerosol light scattering and dew formation affected the carbon exchange at our site, we used an empirical model to analyze the respective influence of these variables on NEE. We modified the Generalized Rectangular Hyperbolae developed by Gu et al. (2002) in order to take into account surface wetness:

$$F_{\text{NEE}} = c_1 \exp(c_2 T_{\text{a}}) - \frac{(\alpha_{\text{f}} I_{\text{f}} + \alpha_{\text{d}} I_{\text{d}})(\beta_{\text{f}} I_{\text{f}} + \beta_{\text{d}} I_{\text{d}})}{(\beta_{\text{f}} I_{\text{f}} + \beta_{\text{d}} I_{\text{d}}) + (\alpha_{\text{f}} I_{\text{f}} + \alpha_{\text{d}} I_{\text{d}}) I_{\text{t}}} \times \left(1 + \frac{1}{W_{\text{s}} - W_0}\right)$$
(2)

where F_{NEE} is net ecosystem exchange flux (µmol m⁻² s⁻¹), T_{a} is air temperature (°C); c_1 , c_2



Fig. 5. Observed and predicted NEE by Eq. (2). The 1:1 relationship is shown.

are parameters; I_t , I_f and I_d are total, diffuse and direct PAR, respectively (µmol m⁻² s⁻¹); β_f , α_f , β_d , α_d are fitted parameters; W_s is surface wetness (%), and W_0 is a fitted parameter. We fitted the parameters of Eq. (2) to our data using a nonlinear procedure (PROC NLIN, SAS Software, SAS Institute Inc., Cary, NC). Mean absolute error (MAE) and root mean square errors (RMSE) were also determined (Janssen and Heuberger, 1995).

We recognized that changing VPD also influences stomatal conductance and GEP, particularly at high VPD values. However, the effects of variations in VPD were not included in this model because adding an additional variable complicates fitting the parameters of the equation. Instead we attempted to exclude any potential effect of low relative humidity on stomatal conductance and GEP by using only data when VPD < 1.5 kPa. The diurnal pattern of NEE for this sub-dataset was similar to the complete dataset, with slightly larger mean CO₂ uptake, especially during the afternoon (Fig. 3b). In the low VPD dataset, the mean difference between morning and afternoon was only 1.0 °C for air temperature and 0.05 kPa for VPD.

Eq. (2) predicts 70% of the variance in the NEE data, with a MAE and RMSE of 1.9 and 2.4 μ mol m⁻² s⁻¹, respectively (Fig. 5). These values are in the range reported in previous studies using similar equations (Gu et al., 2002). Standard errors of the parameters are low and they are all highly significant (Table 2).

Table 2 Parameter values and statistics for the Eq. (2) (n = 1083)

		1	,
Parameter	Value	S.E.	P-value
<i>c</i> ₁	1.35	0.31	0.00
<i>c</i> ₂	0.037	0.0097	0.00
α_{f}	0.020	0.0023	0.00
$\alpha_{\rm d}$	0.011	0.0013	0.00
$\beta_{ m f}$	55.1	14.0	0.00
$\beta_{\rm d}$	24.0	3.5	0.00
W_0	32.0	0.3	0.00

A model sensitivity analysis was performed with Eq. (2) by calculating NEE while independently varying PAR, surface wetness and air temperature (Fig. 6). Response of NEE to temperature had a typical exponential relationship (Fig. 6a) with a $Q_{10} = 1.44$, which is similar to what has been measured at our site based on respiration measurements at ecosystem level $(Q_{10} = 1.6, (Xu \text{ et al.}, 2001))$ (Table 2). Total radiation and the fraction of diffuse PAR positively influenced carbon uptake as shown in previous studies (Baldocchi et al., 1997; Gu et al., 2002; Hollinger et al., 1994) (Fig. 6b). Parameters of the model showed higher light use efficiency ($\alpha_f > \alpha_d$) and higher capacity to resist saturation ($\beta_f > \beta_d$) for diffuse PAR than for direct PAR (Table 2). Similar results were presented by Gu et al. (2002). Coupled to this, sensitivity analysis showed a reduction of CO₂ uptake when wetness increased (Fig. 6c). The effect on NEE was most pronounced when surface wetness was greater than 10%, and thus was mainly restricted to the early morning.

Eq. (2) allows us to separately evaluate the effects of variations in PAR, surface wetness and air temperature on the diurnal pattern of NEE. Three runs were performed by sequentially inputting the observations of PAR, surface wetness and air temperature during the summer 2002, while keeping the other input variables constant at their mean value. Morning (<12:00) and afternoon (>12:00) predicted values were separated for each of these three runs. Sums of NEE were computed and compared for these two periods. The variation of air temperature alone caused mean CO2 uptake by the forest to be 4% smaller in the afternoon than in the morning. Variations of surface wetness suppressed uptake in the morning by 11%. The combination of variations in total PAR (decrease in the afternoon) and the fraction of diffuse PAR (increase in the afternoon) enhanced the CO_2 uptake by 8% in the afternoon.



Fig. 6. Predicted response of NEE with the model 2: (a) sensitivity to air temperature (PAR = 1500 μ mol m⁻² s⁻¹, $I_f/I_t = 0.5$, $W_s = 15\%$), (b) sensitivity to diffuse PAR fraction ($T_a = 18$ °C, $W_1 = 15\%$), (c) sensitivity to surface wetness ($T_a = 18$ °C, PAR = 1500 μ mol m⁻² s⁻¹, $I_f/I_t = 0.5$).

In conclusion, analyses done with Eq. (2) suggest that the enhancement of canopy photosynthesis due to the increase of the fraction of diffuse radiation in the afternoon was larger than the reduction of photosynthesis due to aerosol light extinction. It also suggests that the unusual diurnal pattern of NEE at our site was due both to a suppression of photosynthesis in the morning because of dew formation at the leaf surface, and to an enhancement in photosynthesis in late afternoon because of the positive effect of diffuse PAR.

4. Conclusion

An unusual diurnal pattern of NEE has been consistently observed at the Blodgett Ameriflux site. We present observational evidence of a link between anthropogenic atmospheric aerosols and the uptake of carbon by this ecosystem on a diurnal timescale. We show that anthropogenic aerosols increased the fraction of diffuse radiation during the afternoon, while decreasing the total incoming radiation. Our analysis suggests that these changes resulted in more photosynthetic CO₂ uptake because the positive effect of diffuse radiation was stronger than the negative effect of light extinction. We also presented observational evidence at the canopy scale that morning dew on the leaf surfaces likely reduced canopy photosynthesis, decreasing net carbon uptake by this coniferous ecosystem. These two mechanisms could be especially important at our site because of its Mediterranean mountain climate and its location

downwind of major pollution sources. Recently, Niyogi et al. (2004) reported the first direct observations that the potential of the vegetated land surface to be a sink for atmospheric carbon at regional scales could depend on routine aerosol variability. Assuming that aerosol light scattering and/or surface wetness significantly impacts carbon balance at larger scales, predicted future changes in surface air temperature and aerosol loading should have an impact on carbon sequestration by vegetation globally.

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Appendix A. Symbols and acronyms

$\alpha_{\rm f}$ and $\alpha_{\rm d}$	canopy light use efficiency for
$\beta_{\rm f}$ and $\beta_{\rm d}$	diffuse and direct PAR, respectively capacity to resist saturation for
β	diffuse and direct PAR, respectively solar elevation angle
ER or $F_{\rm ER}$	ecosystem respiration

GEP or F_{GEP}	gross ecosystem productivity
k _t	clearness index
LAI	leaf area index
MAE	mean absolute error
NEE or F_{NEE}	net ecosystem exchange
Р	precipitation
PAR or I_t	total photosynthetically
I_{f}	active radiation diffuse photosynthetically
I _d	active radiation direct photosynthetically
Patms	active radiation atmospheric pressure
PST	pacific standard time
r _h	relative humidity
RMSE	root mean square of error
S	total incoming solar radiation
Se	extraterrestrial solar radiation
$S_{\rm sc}$	solar constant
T_{a}	air temperature
$T_{\rm d}$	day of year
u [*]	friction velocity
VPD	atmospheric vapor pressure deficit
$W_{\rm s}$	surface wetness
W_0	parameter of Eq. (2)

Appendix B. Diffuse radiation model

Diffuse PAR was not measured during summer 2002 concurrently with the aerosol measurements. Using data from April to August 2003, we tested several partitioning models to calculate diffuse and direct PAR. The approach presented in Spitters (1986), coupled with equations from Reindl et al. (1990) as used in Gu et al. (1999, 2002) greatly overestimated the diffuse fraction of PAR (Fig. B.1). The Weiss-Norman model (Weiss and Norman, 1985) was tested as well and gave similar results. These models were mostly developed and tested in temperate regions where relative humidity and cloud cover are typically higher than in Mediterranean regions. Our site in the mountains of California is characterized by a seasonally dry Mediterranean climate: the sky is usually very clear in the morning during the summer, with incoming total irradiance occasionally reaching values $\sim 1000 \text{ W m}^{-2}$.

We built our own model using a similar approach to Reindl et al. (1990). First, the clearness index (k_t) was



Fig. B.1. Observed and predicted diffuse PAR using equations found in Spitters (1986) and Reindl et al. (1990), as used by Gu et al. (1999).

calculated as:

$$k_{\rm t} = \frac{S}{S_{\rm e}} \tag{B.1}$$

where

$$S = 0.45I_{\rm t} \tag{B.2}$$

$$S_{\rm e} = S_{\rm sc} \left[1 + 0.033 \cos \frac{360t_{\rm d}}{365} \right] \sin \beta$$
 (B.3)

where S denotes the total incoming radiation at the Earth's surface (W m⁻²), I_t is the total photosynthetically active radiation measured at the top of the tower (μ mol m⁻² s⁻¹), S_e denotes the extraterrestrial radiation (W m⁻²), S_{sc} is the solar constant (1370 W m⁻²), $T_{\rm d}$ is the day of the year, and β is the solar elevation angle (radians). Eq. (B.2) was determined based on data from summer 2003 ($r^2 = 0.99$, P < 0.001). The solar elevation angle was calculated following the procedure by NOAA based on equations from (Meeus, 1999). The k_t domain was divided into three intervals as presented by Reindl et al. (1990), which were found to be suitable in our case (see below). In each interval, a multiple regression was used to model the fraction of diffuse PAR as a function of several predictors selected by a stepwise regression. The potential predictors included: PAR, clearness index, sine of solar



Fig. B.2. Observed and predicted (a) fraction of diffuse PAR, and (b) diffuse PAR using the model developed in this paper (Eqs. (B.1) to (B.6)). The 1:1 relationship is shown.

elevation angle $(\sin \beta)$, air temperature (T_a) , relative humidity (r_h) , precipitation (P), and atmospheric pressure (P_{atms}) . Based on the results of the regression, the fraction of diffuse PAR can be determined using the following:

Interval:
$$0 \le k_t \le 0.3$$

 $\frac{I_f}{I_t} = 1.0146 - 3.2194 \times 10^{-5} \text{ PAR}$
 $-0.1238k_t - 1.2199T_a$ (B.4)
Interval: $0.3 < k_t < 0.78$
 $\frac{I_f}{I_t} = 3.7340 - 2.9622 \times 10^{-4} \text{ PAR}$
 $-0.8590k_t + 0.5013 \sin \beta - 1.0475T_a$
 $+ 0.2342r_h$ (B.5)
Interval: $k_t \ge 0.78$
 $\frac{I_f}{I_t} = 4.5803 - 4.3043 \text{ PAR} + 2.1435k_t$
 $+ 0.6380 \sin \beta + 5.74 \times 10^{-3}T_a$
 $+ 0.5220r_h + 2.6073P - 7.0565$

$$\times 10^{-3} P_{\rm atms} \tag{B.6}$$

where $I_{\rm f}$ is the diffuse radiation and $I_{\rm t}$ is the total radiation. Fig. B.2 shows the good agreement we

found between observations and predictions of diffuse PAR in summer 2003. The coefficients of determination (r^2) are 0.92 and 0.79 for the diffuse fraction of PAR and the diffuse PAR, respectively. These values as well as MAE and RMSE are in the range reported in other publications (Gu et al., 1999, 2002; Reindl et al., 1990; Spitters, 1986). The regressions calculated between observations and predictions closely follow the 1:1 relationship, with a near zero intercept. We used this model to predict diffuse PAR during summer 2002.

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