VARIATIONS IN SOIL CO_2 CONCENTRATIONS AND ISOTOPIC VALUES IN A SEMI-ARID REGION DUE TO BIOTIC AND ABIOTIC PROCESSES IN THE UNSATURATED ZONE[1]

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ABSTRACT. A study of CO_2 in soil gas was conducted in a bare plot in the unsaturated zone (USZ) of Yatir Forest, northern Negev, Israel. In 2006, 6 tubes for sampling of soil gas were inserted into the USZ to depths of 30, 60, 90, 120, 200, and 240 cm. Profiles of soil gas in the USZ were collected from the tubes 5 times between October 2007 and September 2008. Measurements of the collected profiles of soil gas were of CO_2 (ppm), $\delta^{13}C$ (‰), and $\Delta^{14}C$ (‰). At all times, the concentration of CO_2 in the soil gas was higher than in the air at the surface (CO_2 ~ 400 ppm; $\delta^{13}C$ ~ –9‰). The main source of the CO_2 in soil gas is from biotic activity released through roots of trees and of seasonal plants close to the surface. In the winter, the CO_2 concentrations were lowest (6000 ppm) and the $\delta^{13}C$ was –20‰. In the spring and through the summer, the CO_2 concentration increased. It was estimated that the major source of CO_2 is at ~240 cm depth ($\delta^{13}C$ ~ –22‰; CO_2 ~ 9000 ppm) or below. Above this level, the concentrations decrease and the $\delta^{13}C$ (‰) become more positive. The ^{14}C values in the measured profile are all less than atmospheric and biotic ^{14}C. It was deduced that biotic CO_2 dissolves in porewater to form carbonic acid, which then dissolves secondary carbonate ($\delta^{13}C$ ~ –8‰; ^{14}C ~ –900‰) from the sediments of the USZ. With the ^{14}C data, the subsequent release of CO_2 into the soil gas was then estimated. The ^{14}C data, supported by the ^{13}C and CO_2 data, also indicate a biotic source at the root zone, at about 90 cm depth.

INTRODUCTION

CO_2 from the unsaturated zone (USZ) is considered to play a significant role in the balance of this gas in the atmosphere. Therefore, a significant effort has been expended in determining the concentration and isotopic composition of CO_2 in the USZ and in understanding the effect that it has on the atmosphere. Most of the work done was by collecting the respired CO_2 in an isolated volume above the USZ. Davidson (1995) estimated that the $\delta^{13}C$ (‰) of the CO_2 in the soil gas came mostly from C_3 sources.

Parker et al. (1983) studied the respiration of CO_2 above grassland in Texas and estimated the flux of CO_2 out of the USZ and the rate of mineralization in the USZ. Gorzyca et al. (these proceedings) studied soil-respired CO_2 and its isotopes for 7 yr in several ecosystems in southern Poland by collection of CO_2 above the USZ. They measured the carbon isotopes and found a dependence on industrialization in the vicinity of their sampling sites.

In the USZ, Pumpanen et al. (2008) installed permanent probes in the USZ to collect data on the profile of CO_2 concentrations in the USZ in spring time. They applied diffusion to calculate the efflux from the soil to the atmosphere for that time. Liu et al. (2006) used CO_2 mass balance and $\delta^{13}C$ to calculate the proportions of CO_2 from decomposed organic matter and from air in the soil gas of a boreal forest. They studied the transport of CO_2 in soil gas with ^{14}C. The contribution of CO_2 from carbonate mineral in the USZ is considered to be significant. According to Deines et al. (1974), dissolution of carbonate minerals in the USZ is regulated by acidity and pCO_2, with higher pCO_2 resulting in greater amounts of carbonate dissolution.

[1]This paper is dedicated to the memory of Prof JR Gat (1926–2012), mentor and a good friend.

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The purpose of this study was to understand processes in the USZ in a bare plot, the various sources of CO_2 therein, and the mode of its transfer to and from the atmosphere. For this purpose, a matrix of 5 profiles of CO_2 and $\delta^{13}CO_2$ was obtained over 1 yr (October 2007 to September 2008). ^{14}C was measured in the profile of May 2008.

SITE DESCRIPTION

The Yatir Forest (31°20′N, 35°03′E, 650 m asl) is located in a semi-arid zone (290 mm/yr rain) on the southwestern flanks of the Judea Mountains of Israel (Figure 1a). The forest was planted by the Jewish National Fund in the 1960s and it is the site of the Yatir bio-ecological station of the Weizmann Institute of Science, which was erected in spring 2000 (Grünzweig et al. 2003). Among the many studies done at that station are those of CO_2 efflux from the USZ with diurnal resolution, reported by Maseyk et al. (2008) and Grünzweig et al. (2009). The site of the present study of soil gas CO_2 is a bare 20 × 20 m plot in the forest, 100 m to the east of the Yatir bio-ecological station.

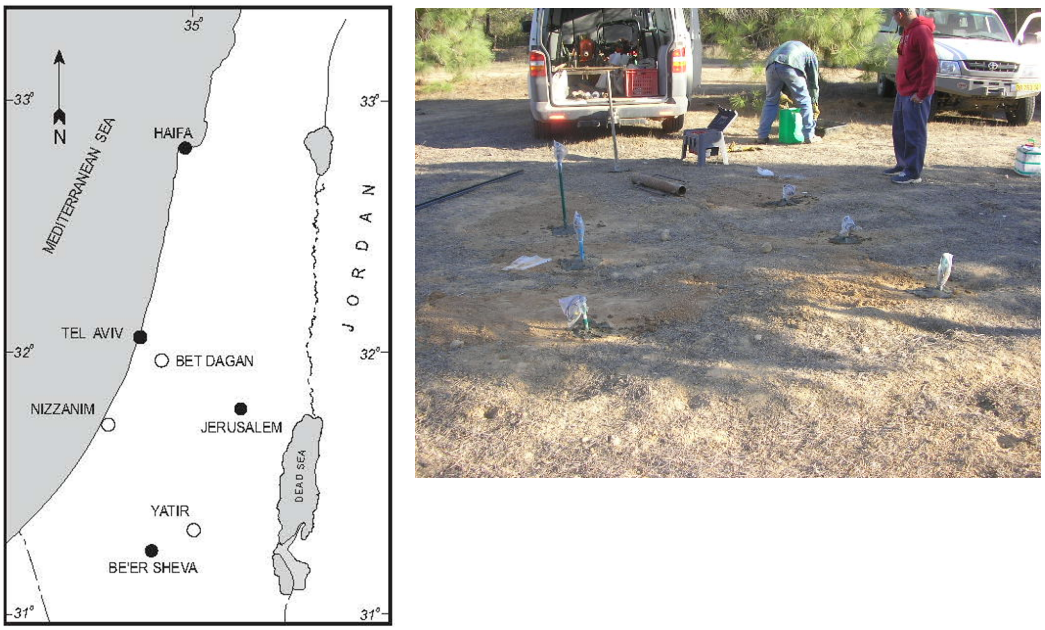

Figure 1 (a, left) Map showing the location of Yatir, Nizzanim, and Bet Dagan; (b, right) Layout of the soil gas tubes in Yatir.

The USZ in the Yatir Forest is composed of imported loess and has no relation to the underlying carbonate rock. Its thickness in most of the forest is ~1 m, but in the bare plot used in the present study the thickness is >4.5 m. The organic carbon content of the soil is 1.3% at 1.8 m, which is much less than the 9–23% reported by Liu et al. (2006) for a forest in Japan.

METHODS

In December 2006, 6 holes were drilled into the USZ of a bare plot in the Yatir Forest (Figure 1b) for the collection of soil gas. The holes were drilled to depths of 30, 60, 90, 120, 200, and 240 cm. PVC tubes (18 mm diameter) were inserted into the holes and left open at both ends: one in the USZ to the specified depth, and the other to the atmosphere. The system was left to settle for 10 months before the sampling began. Between the samplings, the local fauna sometimes gnawed the tops of

the sampling tubes, but, when this happened, the problems were overcome successfully. After the third sampling, the 90-cm-deep tube became blocked and could not be further sampled.

Two methods of sampling were used and gave similar results. In the first method (Carmi 2007), calibrated glass vessels fitted with valves were pre-evacuated in the lab at the Kimmel Center for Archaeological Research of the Weizmann Institute of Science, and then connected to the tubes in the field. The valves were then opened and the pressure in them was monitored. In the USZ, the pressure of soil gas is atmospheric. Following several minutes for equilibration, the valves were closed. In the lab, the vessels were connected to a vacuum line, the volume of soil gas was measured, and the CO_2 was separated from the soil gas. The volume of CO_2 was then measured and its concentration (ppm) was calculated (Carmi 2007). The CO_2 was saved for further measurements. ^{13}C was measured in a Finnigan Mat 250 mass spectrometer in the Department of Environmental Science and Energy Research at the Weizmann Institute of Science. ^{14}C was measured in the profile of May 2008 by the NSF-Arizona AMS laboratory. In the second method of sampling, vessels with 2 valves were brought to the field, connected to the tubes, flushed 8–10 times by pumping soil gas through them after which the valves were closed. In the lab, the vessels were connected directly to a Carlo Erba 1108 elemental analyzer coupled to a continuous-flow Micromass isotope ratio spectrometer, for measurement of the concentrations of CO_2 (ppm) and $\delta^{13}C$ (‰) (Klein et al. 2005). In this method, the samples were small and ^{14}C was not measured.

RESULTS AND DISCUSSION

The results of the 5 samplings at 6 depths in the USZ of the bare plot near the Yatir forest are given in Table 1. The data are presented graphically in 2 ways: as depth profiles at the time of sampling (Figures 2a,b and c) as well as chronologically for each depth (Figure 3a,b).

Table 1 Soil gas data from Yatir.

	10/2007		1/2008		5/2008			8/2008		9/2008	
Depth (cm)	$\delta^{13}C$ (‰)	CO_2 (ppm)	$\delta^{13}C$ (‰)	CO_2 (ppm)	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)	CO_2 (ppm)	$\delta^{13}C$ (‰)	CO_2 (ppm)	$\delta^{13}C$ (‰)	CO_2 (ppm)
30	–16.3	715	–11.6	744	–247.5	–15.4	1372	–16.7	2980	–17.9	1730
60		690	–16.4	747	–148.9	–17.4		–16.4	2298	–17.4	1230
90	–17.6	2000	–16.9	1516	–101.9	–17.8	4732				
120		2095	–16.8	2052	–144.2	–17.1		–17.1	4052	–17.7	2538
200	–17.9	5040	–16.3	1604	–185.1	–14.8	4912	–18.6	6238	–19.4	4715
240			–19.9	6021			8637	–21.0	9694	–19.9	7742

Depth Profiles of CO_2 and Its Isotopes

The concentration of CO_2 (ppm) in the soil gas of Yatir (Figure 2a and Table 1) is always highest at the 240 cm depth, suggesting a nearby source. From 240 cm upwards towards the surface, CO_2 generally decreases. There are a few exceptions to this trend, such as in August and September 2008 when there was, near the surface (at ~30 cm depth), an increase in the CO_2 concentration relative to earlier profiles. At the root zone, 90 cm deep, an apparent increase in the CO_2 concentration is observed in May 2008.

The most negative values of $\delta^{13}C$ at the bottom of the profiles (August and September 2008) are quite similar to those of C_3 plants. This suggests that the sources are roots of C_3 plants in the near vicinity (Figure 2b and Table 1). The $\delta^{13}C$ values become more positive towards the top of the USZ. The most positive $\delta^{13}C$ is observed at the top of the section in January 2008. The low CO_2 and the

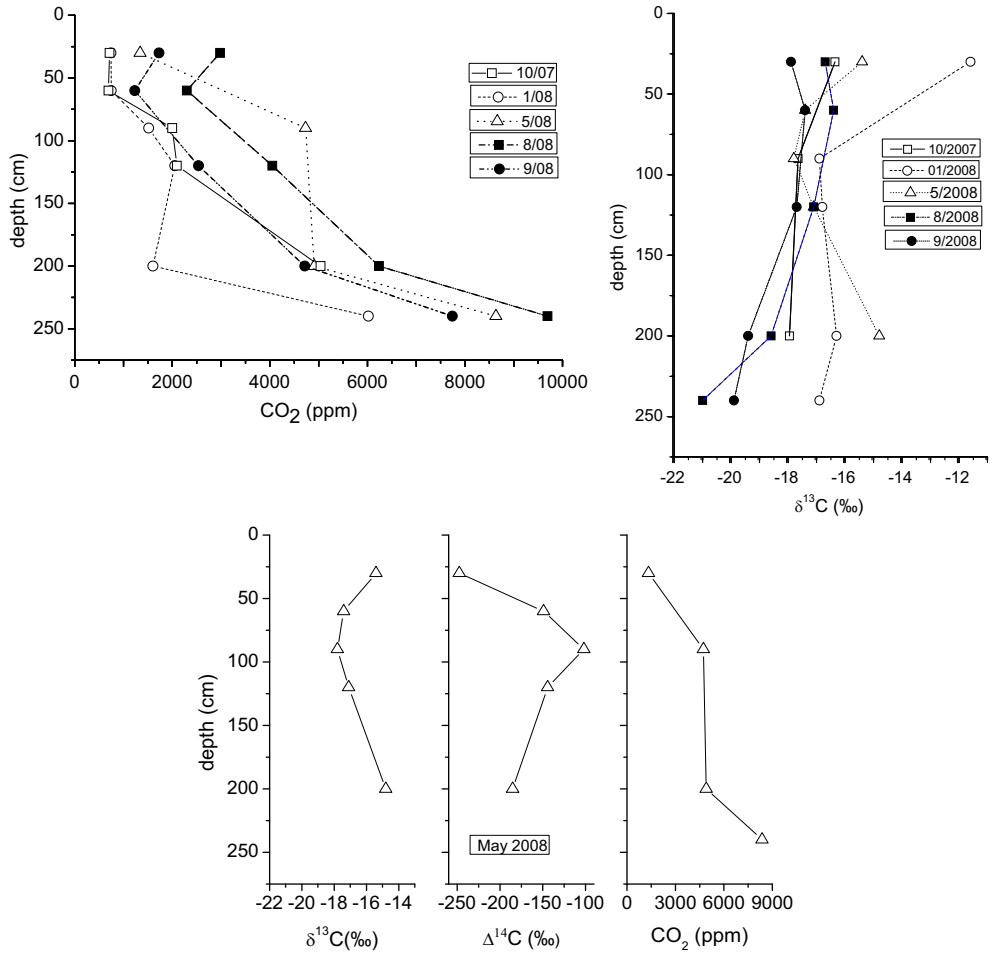

Figure 2 Depth profiles of Yatir soil gas at different times: (a, top left) CO_2 (ppm); (b, top right) $\delta^{13}C$ (‰); and (c, bottom) CO_2 (ppm), $\delta^{13}C$ (‰) and $\Delta^{14}C$ (‰) in May 2008.

more positive $\delta^{13}C$ of January 2008 at the top of the profile suggest an input from the atmosphere above (400 ppm and –9‰). At the end of the summer, the increased concentration of CO_2 at the top of the section together with the slightly more negative $\delta^{13}C$ suggests contribution from seasonal plants. The dry remains of seasonal plants are seen on the surface in December (Figure 1b).

The profiles of CO_2, $\delta^{13}C$ (‰), and $\Delta^{14}C$ (‰) in May 2008 are shown in Figure 2c and Table 1. The maximum in the $\Delta^{14}C$ (‰) profile is at 90 cm depth. Taken together with the CO_2 and $\delta^{13}C$ data, the $\Delta^{14}C$ (‰) clearly suggests a local biotic source of CO_2 well within the root zone. The $\Delta^{14}C$ (‰) values in the profile are all less than the measured atmospheric values of 49.5‰ in 2008 (Levin et al. 2010) and less than the $\Delta^{14}C$ in the biosphere, which is assumed to be similar to the atmospheric $\Delta^{14}C$. The overall low $\Delta^{14}C$ in the soil gas CO_2 requires a contribution from an additional source with a low ^{14}C concentration.

Seasonal Variations of CO_2 and $\delta^{13}C$

An annual cycle in the CO_2 concentrations is quite clearly manifested. In the fall and winter, the concentrations of CO_2 (ppm) decrease at all depths (Figure 3a and Table 1). This is probably due to reduced production of CO_2 by the deep source. However, at the top of the section (30 cm), the rate of decrease is smaller due to production of CO_2 in the seasonal sources at the top and at the 90-cm-deep source. In the spring and summer, the concentrations increase at all depths, at a rather similar rate. Towards the fall (September 2008), the concentrations decrease again towards dormancy.

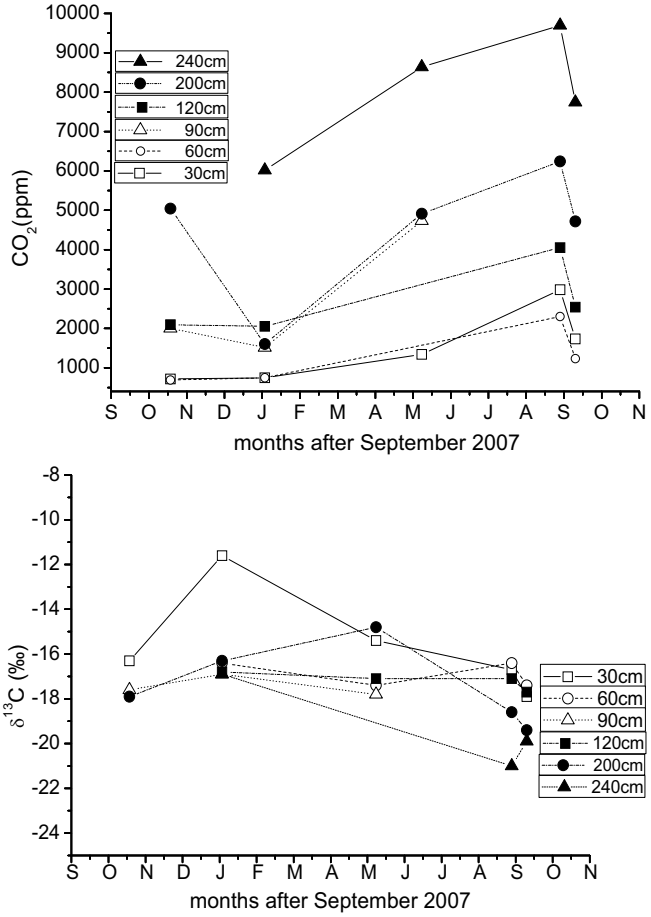

Figure 3 Seasonal variations of Yatir soil gas at different depths: (a, top) CO_2 (ppm); (b, bottom) $\delta^{13}C$ (‰).

The $\delta^{13}C$ (Figure 3b) at all depths and along the annual cycle seems to converge near –16 to –18‰. There are 2 noteworthy exceptions: one occurs at the top in January 2008, which displays the least negative value and is closest to that of the air above the USZ (as discussed above). The other exception is at 240 cm depth where the $\delta^{13}C$ is always more negative and thus closest to that of C_3 plants.

Sources of CO_2 in the Soil Gas

Three biotic sources are suggested by the data at 2 depths in the May 2008 profile (90 and 240 cm perennial roots and seasonal plant roots at the top) and 1 abiotic source (air present in the soil gas of the USZ). However, as shown in Figure 2c, the $\Delta^{14}C$ in the soil gas CO_2 (–102‰ to –248‰, Table 1) is less than the atmospheric and biosphere level of 49.5‰ in 2008 (Levin et al. 2010) (Table 1 and Figure 2c). These $\Delta^{14}C$ (‰) values require an additional (fifth) source that contributes CO_2 with low ^{14}C to the soil gas. The likeliest candidate for this source is the carbonate in the sediments of Yatir, a second abiotic source of CO_2 in the soil gas. $\Delta^{14}C$ measured in the carbonate of the sediments of Yatir was found to be –899‰ to –986‰ (Table 2), while the associated $\delta^{13}CO_2$ was –8‰. Deines et al. (1974) have shown that in the presence of water and high concentrations of CO_2, dissolution of sediments can occur and contribute CO_2 to the soil gas. Assuming this to be the case at Yatir, this dissolution was estimated for May 2008 at 2 depths in the profile (90 and 200 cm). The following rough approximation was used:

$$\Delta^{14}C \times C = \Delta^{14}C_s \times C_s + \Delta^{14}C_a \times (C_b + C_a) \quad (1)$$

and

$$C = C_s + C_b + C_a \quad (2)$$

The first equation describes the mass balance for $^{14}CO_2$. The subscripts *s*, *b*, and *a* are, respectively, the sediment, biosphere, and atmosphere. *C* is the total concentration of CO_2 in soil gas and C_s, C_b, and C_a are the respective CO_2 contributions from abiotic and biotic sources in the sediment and from atmospheric origin. $\Delta^{14}C_a = \Delta^{14}C_b = 49.5‰$ is the assumed value for the CO_2 in the atmosphere and the biosphere. The second equation is the mass balance for CO_2. The results of the calculations for 2 levels in the USZ are shown in Table 3 and Figure 4. It is evident that the contribution from the sediment, C_s, is significant, in the range of 16–22% and it is believed that this may represent the profile as a whole.

Table 2 Sources of CO_2 in the soil gas of the USZ of Yatir in May 2008 calculated using Equations 1 and 2 (see above). In every cell of the table, the first line is the actual data and the second line (in *italics*) is the same data in percent (%) (*s* = sediment; *b* = biosphere; *a* = air).

Depth (cm)	C^{i} (ppm) (%)	$^{14}C^{i}$ (‰)	C_s^{ii} (ppm) (%)	$^{14}C_s^{i}$ (‰)	C_b^{i} (ppm) (%)	$^{14}C_b^{i}$ (‰)	C_a^{iii} (ppm) (%)	$^{14}C_a^{i}$ (‰)
90	4730	–101.9	754	–899	3576	49.5	400	49.5
	100		*15.9*		*75.6*		*8.5*	
200	4912	–185.1	1113	–986	3399	49.5	400	49.5
	100		*22.7*		*69.2*		*8.1*	

[i] Measured data.
[ii] Results of calculations.
[iii] Assumed.

In principle, the $\delta^{13}C$ values could be used for a similar estimate of the contribution of CO_2 by dissolution from the sediment. However, unlike the ^{14}C values in soil gas CO_2 and in CO_2 evolved from sedimentary carbonate, which are very different (Table 2), the respective $\delta^{13}C$ values are very close. The sedimentary carbonate has a $\delta^{13}C = –8.0‰$; following Clark and Fritz (1997), the $\delta^{13}C$ of the released CO_2 would be –17.2‰. The soil gas CO_2 ranges between –14.8 and –17.8‰ and the atmospheric and biotic values are also not very different. Therefore, the difference in the $\delta^{13}C$ values is too small to provide a reasonable estimate for the sedimentary contribution.

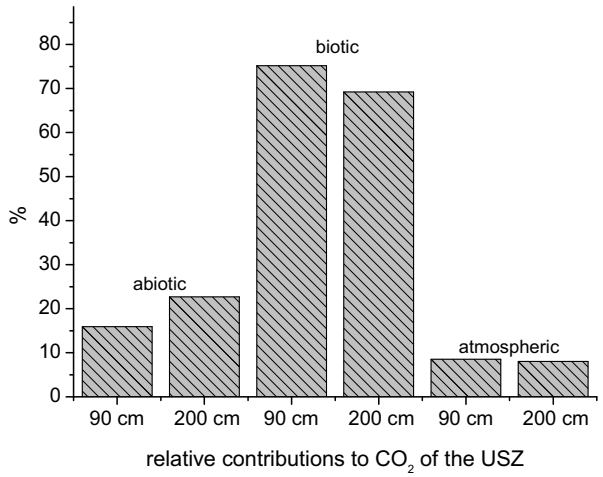

Figure 4 Relative biotic, abiotic, and atmospheric contributions to the CO_2 in the soil gas of the USZ in Yatir.

Correlations Between CO_2 and $\delta^{13}C$

Figures 5a–e show the correlations between CO_2 (ppm) and $\delta^{13}C$ (‰) for the 5 profiles of soil gas in Yatir. The correlation in October (Figure 5a) seems quite good and suggests a dependence between the 2 variables: $\delta^{13}C$ (‰) more negative with higher concentration of CO_2 and *vice versa*. In January (Figure 5b), the dependence is much less pronounced, but the excursion towards less negative $\delta^{13}C$ (‰) at the top again suggests influx from the air above the USZ. Not much can be said about the correlation in May (Figure 5c) because the CO_2 data is incomplete (Table 1).

The correlations in August (Figure 5d) and September (Figure 5e) are good (R^2 = 0.99 and 0.91, respectively) and the relations that describe them are

$$\delta^{13}C\ (‰) = -0.0006 \times CO_2 - 14.77 \quad (3)$$

and

$$\delta^{13}C\ (‰) = -0.0004 \times CO_2 - 17.03 \quad (4)$$

Extrapolations to the atmosphere (400 ppm CO_2) give $\delta^{13}C$ = –15‰ in August and $\delta^{13}C$ = –17.2‰ in September. However, the 2 values are significantly more negative than the atmospheric value of $\delta^{13}C$ = –9‰, implying that there is a significant flow of CO_2 from the USZ to the atmosphere in these months (see below).

Fluxes of CO_2 Through the Interface Between the USZ and the Atmosphere

The data presented suggest that transport of CO_2 by diffusion is an important process in the USZ. Indeed, outflow from the USZ to the atmosphere by diffusion has been suggested by Cerling (1984). Influx of CO_2 from the atmosphere into the USZ, characterized by a lower concentration of CO_2 and less negative $\delta^{13}C$ (400 ppm and –9‰, respectively) is responsible for the gradual dilution of the concentration of CO_2 in the USZ, a larger dilution at the top decreasing downward (Figure 2a). This is most evident in January 2008 (Figure 2b). It is possible to estimate the flux using the Fick equation:

$$F = -r\frac{dC}{dz} \quad (5)$$

where F is the flux of CO_2 in g C m^{-2} $month^{-1}$, C is the concentration of CO_2 (g CO_2 m^{-3}), z is the depth in m, and r is the effective diffusion coefficient:

$$r = D \times p \times t \quad (6)$$

where D is the diffusion coefficient of CO_2 in air (37.9 m^2 $month^{-1}$), p is the porosity (0.51), and t is the tortuosity (0.6) (Cerling 1984). For the site of Yatir, r = 11.6 m^2 $month^{-1}$. Since both the outflux and influx proceed simultaneously, the result of the calculation will be a net flux.

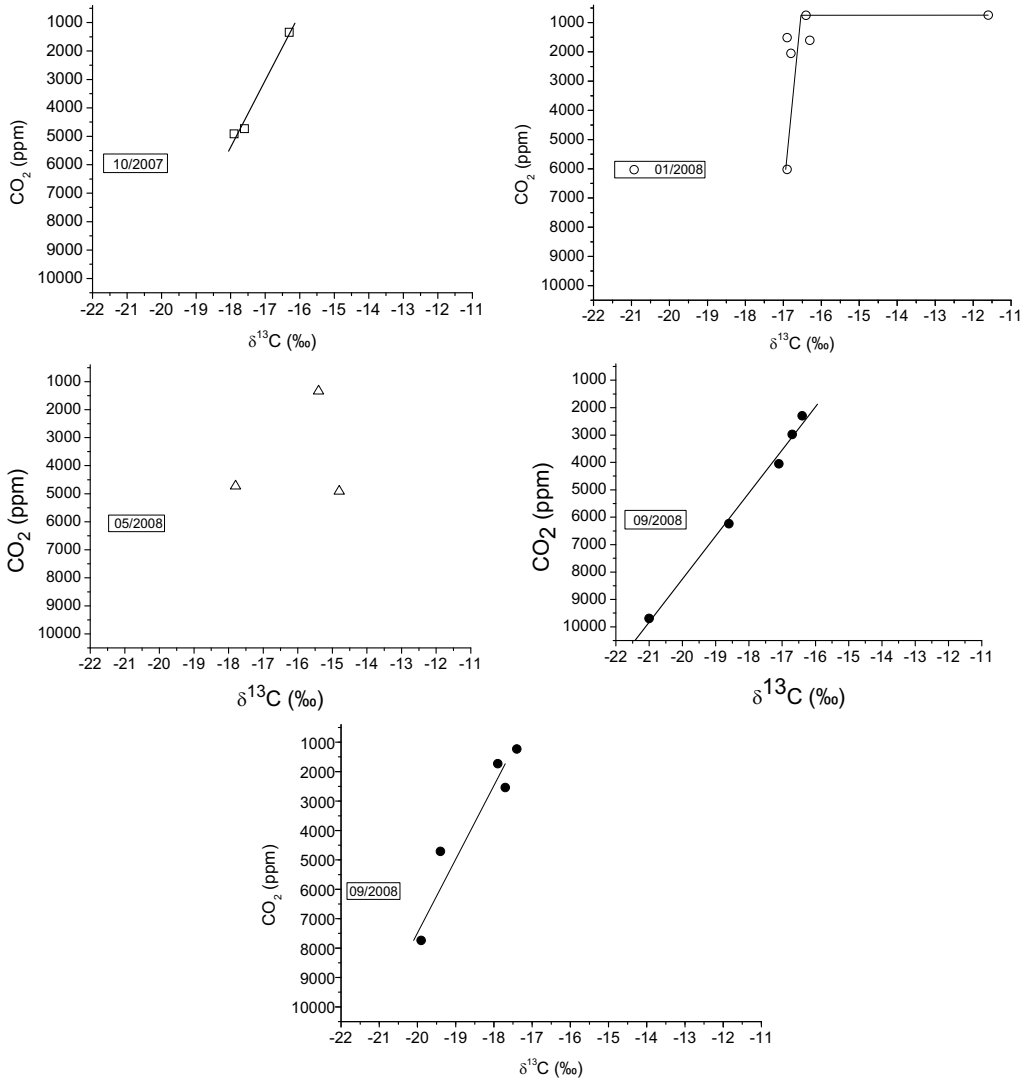

Figure 5 Correlations between CO_2 (ppm) and $\delta^{13}C$ (‰) in Yatir soil gas: (a, top left) October 2007; (b, top right) January 2008; (c, middle left) May 2008; (d, middle right) August 2008; (e, bottom) September 2008.

Figure 6 shows the calculated monthly net outfluxes of CO_2 from the USZ at the Yatir bare plot calculated by the Fick equation. Data between the months of May and September is not available and thus was estimated by interpolation. The resulting integrated annual outflux is 260 g C m^{-2} yr^{-1}. It

is, in fact, the balance between the fluxes of soil CO_2 from the USZ to the atmosphere and of atmospheric CO_2 to the USZ. This value from the bare plot can be compared to the annual flux in the Yatir Forest itself, with its 300 trees per hectare, which is 406 g C m^{-2} yr^{-1} (Maseyk et al. 2008) (Figure 7a). The source of CO_2 in the USZ is predominantly emanation from underground roots. There are no trees in the bare plot of the present study (Figure 1b); therefore, the density of the roots is low. Emanation of CO_2 is therefore slower and the flux out of the USZ is smaller.

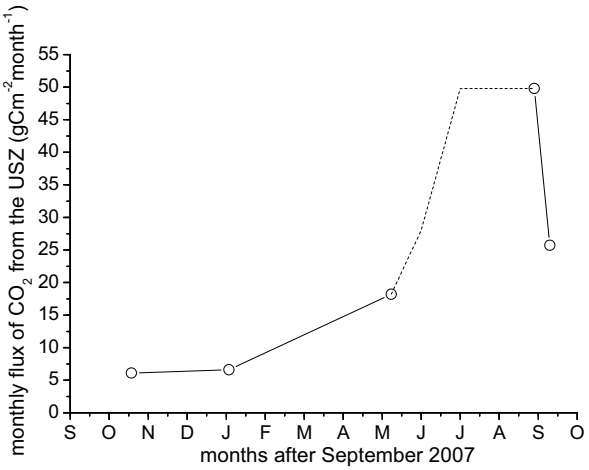

Figure 6 Calculated monthly fluxes of CO_2 (g C m^{-2} $month^{-1}$) from the USZ to the atmosphere in Yatir. The dashed line is an interpolation.

It is of interest to compare the monthly fluxes from the USZ in the Yatir bare plot with plots bare of trees in the more fertile Coastal Plain of Israel (Figure 1a, Table 3, Figure 7b). Here, the porosity is lower (0.35 compared to 0.51 in Yatir) because the proportion of clay in the sediments is much higher than in Yatir. The effective diffusion coefficient r = 7.9 m^2 $month^{-1}$ is thus significantly smaller than at Yatir. However, due to the higher productivity at Nizzanim, and despite the smaller effective diffusion coefficient, the outflux from the bare area of Nizzanim is higher than from the Yatir bare plot, both in winter and in spring (Figure 7b). It is worth noting that the relative increase between the 2 seasons is similar at both sites. In a second bare plot of the Coastal Plain, at the site of the Meteorological Service of Israel at Bet Dagan, the outflux in the spring is less than at the other 2 sites. This smaller outflux of CO_2 seems reasonable because it has been measured next to the meteorological monitoring instruments that are located in a plot of ground that is intentionally kept devoid of vegetation.

Table 3 Comparison of monthly CO_2 outfluxes in 3 sites in Israel calculated from data in Carmi et al. (2009) and Carmi (2007).

Site	Month	Flux (g C m^{-2} $month^{-1}$)
Yatir	February	10.0
Nizzanim	February	15.6
Yatir	May	12.5
Nizzanim	May	18.6
Bet Dagan (Met Service)	May	9.9

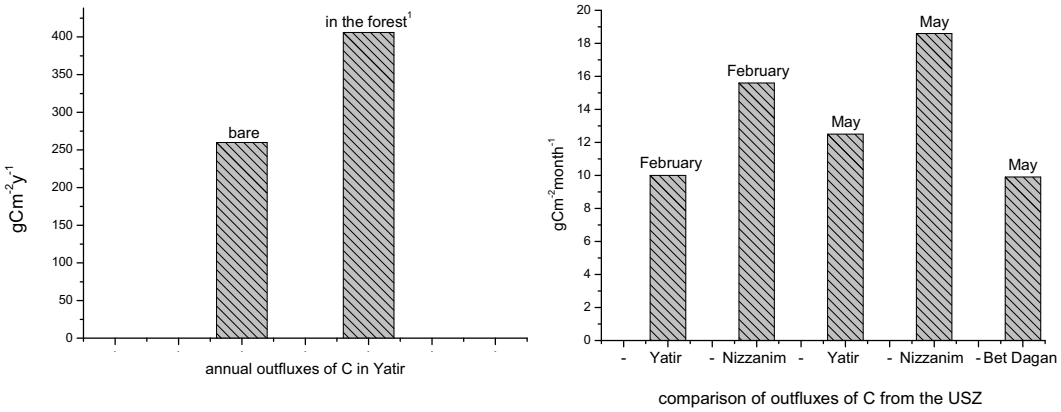

Figure 7 (a, left) Annual outfluxes of carbon in the bare plot and in the forest of Yatir; (b, right) Comparison of monthly outfluxes of carbon in Yatir and in Nizzanim and Bet Dagan in the Coastal Plain of Israel.

CONCLUSIONS

The sampling strategy used in this study was, of necessity, snapshots at discreet times. Yet, the methodology used in this study for measuring the soil CO_2 profiles in the USZ of the bare plot in Yatir garnered several results and conclusions. The sources of CO_2 in the soil gas could be identified as biotic activity of major, minor, and temporary roots of plants, as abiotic dissolution, and atmospheric air. The soil CO_2 is very dynamic, with annual cycles of biotic activity shown by similar seasonal variations in all depths. The outward diffusive fluxes of CO_2 are seasonally variable and are smaller in bare plots than in productive ones. During low biotic productivity (January 2008), $\delta^{13}C$ provided evidence for the flux of CO_2 from the atmosphere into the USZ.

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