

Photo- and thermal degradation in an arid ecosystem

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The role of photo- and thermal degradation for CO₂ and CO fluxes in an arid ecosystem

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Abstract

Recent studies have suggested the potential importance of abiotic degradation in arid ecosystems. In this study, the role of photo- and thermal degradation in ecosystem CO₂ and CO exchange is assessed. A field experiment was performed in Italy using a FTIR-spectrometer coupled to a flux gradient system and to flux chambers. In a laboratory experiment, field samples were exposed to different temperatures and radiation intensities.

No photodegradation-induced CO₂ and CO fluxes were found in the field and in the laboratory study. In the laboratory, thermal degradation fluxes for CO₂ and CO have been observed. In the field, CO uptake and emission have been observed and are proposed to be a result of biological uptake and abiotic thermal degradation-production.

We suggest that previous studies, studying direct photodegradation, have overestimated the role of photodegradation and observed fluxes might be due to thermal degradation, which is an indirect effect of radiation. The potential importance of abiotic decomposition in the form of thermal degradation, especially for arid regions, should be considered in future studies.

1 Introduction

CO₂ is the main carbon species being exchanged between biosphere and atmosphere and the most important anthropogenic greenhouse gas. CO is a less abundant non-greenhouse gas but considered important in the climate debate due to its oxidation process with atmospheric OH⁻ (Stocker et al., 2013). Yearly, terrestrial ecosystems exchange approximately 120 Pg_{yr}⁻¹ of carbon with the atmosphere (Stocker et al., 2013).

Arid ecosystems account for approximately 40% of land area and 20% of the soil carbon pool but are still an unknown factor in climate models (Lal, 2004). In recent studies, the possible importance of abiotic degradation for arid regions, such as photo-

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and thermal degradation, has been recognized (Rutledge et al., 2010; King et al., 2012; Austin and Vivanco, 2006).

1.1 Ecosystem CO₂ fluxes; photo- and thermal degradation

Photodegradation is the direct breakdown of organic matter by radiation. Photodegradation is known to be an important pathway in aquatic ecosystems (Zepp et al., 1998). Recently, the possible importance of photodegradation in terrestrial ecosystems has been suggested (Brandt et al., 2010; Rutledge et al., 2010; Friedlingstein et al., 2006; Austin and Vivanco, 2006). Photodegradation can play an important role in arid ecosystems, where microbial decomposition is restricted (Austin and Vivanco, 2006; Brandt et al., 2010; Lee et al., 2012; Throop and Archer, 2009; Lin and King, 2014). Rutledge (2010) estimated that in arid ecosystems, 19% of the annual CO₂ flux is induced by photodegradation and, in dry summer conditions, even 92% of daytime CO₂ emissions can be attributed to this process.

Photodegradation is attributed to as well UV radiation as to visible radiation (Brandt et al., 2010; Austin and Vivanco, 2006; Bruhn et al., 2009). The biochemical mechanisms behind photodegradation-induced carbon fluxes are not clear; it is proposed that solar radiative energy breaks down the bonds of carboxyl, directly producing CO₂ and other gas species (Lee et al., 2012). Assumed is that rates of photodegradation are dependent on plant and litter tissue type: lignin, the most recalcitrant tissue in plant material (to microbial decomposition) is expected to be most sensitive to photodegradation (Austin and Ballaré, 2010; King et al., 2012). However, while studies reporting photodegradation are multiple, recent studies, aiming to further investigate the process, were unable to observe photodegradation-induced carbon fluxes (Lambie et al., 2014; Uselman et al., 2011; Kirschbaum et al., 2011). A reason for this discrepancy has not yet been found (Kirschbaum et al., 2011; Uselman et al., 2011; Lambie et al., 2014; Throop and Archer, 2007). It is important to notice that in literature, the term photodegradation is sometimes also used for the indirect effects of radiation on decomposition. For a review on studies done on photodegradation, please see King (2012).

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A less studied abiotic degradation pathway is thermal degradation, the temperature dependent degradation of carbon in absence of radiation and possibly oxygen (Lee et al., 2012; Schade et al., 1999; Derendorp et al., 2011). However, photodegradation is considered the more dominant abiotic CO₂ producing process (Lee et al., 2012).

Besides CO₂, also CO and CH₄ are reported as products of photo- and thermal degradation (Schade et al., 1999; Lee et al., 2012; Tarr et al., 1995; Derendorp et al., 2011; Vigano et al., 2008).

1.2 Ecosystem CO fluxes; photo- and thermal degradation

The role of CO in soils and ecosystems is not well understood. Soils are known for being as well a source as a sink for CO (Conrad, 1996). Most likely, the main cause for soil CO uptake is the oxidation of CO to CO₂ or CH₄ by soil bacteria or soil enzymes (Ingersoll et al., 1974; Conrad, 1996; Spratt and Hubbard, 1981; Yonemura et al., 2000; Whalen and Reeburgh, 2001; Bartholomew and Alexander, 1979). Soil CO consumption is found to be dependent on atmospheric CO concentrations and the consumption rate is usually expressed in deposition velocity: the uptake rate divided by the CO concentration (Conrad and Seiler, 1982; Kisselle et al., 2002).

Soil CO emissions have also been reported and are thought to be of non-biological origin (Conrad and Seiler, 1980, 1982). For example, soil CO emissions were found in peatlands (Funk et al., 1994) and in arid soils (Conrad and Seiler, 1982). Living plants are also known to emit a small amount of CO (Kirchhoff et al., 1990; Bruhn et al., 2013; Tarr et al., 1995). However, senescent plant material has been shown to emit 5 to 10 times more (Tarr et al., 1995; Schade et al., 1999; Derendorp et al., 2011); these fluxes, mostly determined in laboratory studies, were attributed to thermal degradation and, to a larger extent, to photodegradation (Derendorp et al., 2011; Lee et al., 2012; Schade et al., 1999).

1.3 Measurement of photo- and thermal degradation

Studying photodegradation is difficult due to the multiple (indirect) effects radiation has on total (biological) decomposition. For example, UV radiation is known to inhibit microbial processes, to change (senescent) tissue chemistry and to shift microbial and fungal communities, thereby affecting microbial decomposition rates in both directions (Uselman et al., 2011; Formánek et al., 2014; Zepp et al., 1998; Williamson et al., 1997). Differentiating photodegradation-induced fluxes from biological sources in field experiments can be achieved by comparison of different flux measurement techniques such as Eddy Covariance (EC) measurements vs. flux chamber measurements and/or soil gradient measurements, in where one method does not receive solar radiation (Rutledge et al., 2010). This approach requires that the areas sensed by the different techniques (footprint) are fully homogeneous, which is not often the case and hard to validate. To study photodegradation, also flux chamber systems can be used, each with a different amount of radiation exposure (Lee et al., 2012; Brandt et al., 2010; Lin and King, 2014).

Studying the role of thermal degradation-induced carbon fluxes is challenging, especially for CO₂ due to the accompanying effect temperature has on microbial decomposition. To study thermal degradation-induced CO₂ production, microbial decomposition should be absent, which can only be achieved in laboratory studies (Lee et al., 2012).

Previous field studies on the role of (in)direct abiotic degradation report very contrasting results (King et al., 2012; Lambie et al., 2014; Rutledge et al., 2010; Kirschbaum et al., 2011; Uselman et al., 2011; Lee et al., 2012). More specific studies are thus needed to better understand this process and its role in the carbon cycle. In this study, we present the results of field and laboratory measurements aimed to evaluate the role of direct photodegradation and thermal degradation in an arid ecosystem.

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2 Materials and methods

2.1 Study site

We performed a field experiment in a grassland (IT-Ro4, harvested cropland, approximately 250 m by 450 m, lat 42.37° N, long 11.92° E, 147 m a.s.l.), in the province of Viterbo, Italy. The climate is Mediterranean, with a typical drought period covering approximately 2 months during summer (July–August). Mean annual temperature is 14 °C and annual rainfall is 755 mm. Such climatic characteristics make the site suitable for abiotic degradation studies. The underlying material is Tuff, soil texture is clay loam and soils are classified as Eutric Cambisol. Yearly, the fieldsite is ploughed to a depth of 20 or 50 cm. Just before the experiment, oat and vetch were cultivated. During the experiment, vegetation was not managed and was a mix of invasive species such as *Amaranthus retroflexus*, *Chenopodium* spp., *Conyza Canadensis*, *Artemisia vulgaris*, *Cirsium* spp., *Mercurialis annua* and *Polygonum* spp. The field study was conducted in July–September 2013. At the beginning of the experiment, most vegetation was dried out, however, patches of active vegetation were observed. Temperature and rainfall during measurements were representative for the period (hot and dry), however, the preceding spring had been cold and rainy in respect to the average.

IT-Ro4 is an experimental site managed by the University of Tuscia (Viterbo). Continuous EC measurements of scalars and energy fluxes are performed (LI-7500 open path analyzer, Licor, Lincoln, Nebraska, USA; Windmaster Pro sonic anemometer, Gill, Hampshire, UK) along with meteorological and environmental measurements (CNR-1, Kipp & Zonen, Delft, the Netherlands; soil water content, CS616, Campbell Scientific, North Logan, USA; soil temperature, CS107, Campbell scientific, North Logan, USA; soil heat flux, HFT3 Soil Heat Flux Plate, Campbell scientific, North Logan, USA).

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2.2 Instrumentation and set up

The analyzer used in this study is based on a Fourier Transform Infrared (FTIR)-spectrometer (Spectronus, Ecotech), for details on the FTIR-analyzer, see Griffith (2012). A FTIR is capable of measuring air concentrations of CO₂, CH₄, N₂O, CO and $\delta^{13}\text{CO}_2$ simultaneously. Before being measured, air samples were dried by a nafion dryer and by a column of magnesium perchlorate. Measurements were corrected for pressure and temperature fluctuations and for cross-sensitivities (Hammer et al., 2013). Background measurements and a calibration routine using two standard gas cylinders were performed weekly. We designed an external manifold box which allowed us to connect the FTIR to a flux gradient (FG) setup and to 2 flux chambers (FC), simultaneously. Both methods provide air concentration data as well as flux data. In this paper, only CO₂ and CO flux data are presented.

2.3 Concentration and flux measurements

FG measurements were performed once per hour. Air inlet heights were at 1.3 and 4.2 m. Air was sampled at 1 L min⁻¹. Sampling lines of stainless steel were used for the experiment. For 30 min h⁻¹, the airflows were led to air sampling bags, after that the bag inlet was closed until analysis. Before the analysis, the FTIR measurement cell was evacuated and flushed twice with measurement air before being filled. Per air sample, a 3 min-spectra (static) measurement was taken. FG measurements were performed at the same point as of the EC set-up (measurement height at 3.5 m). By FG method, fluxes can be calculated by:

$$F = K \frac{\delta C}{\delta z} \quad (1)$$

wherein δC is the difference in concentration of a gas species (mol m⁻³) between the two inlet-heights (δz (m)) and K is the diffusion coefficient (m² s⁻¹), and F the flux (mol m⁻² s⁻¹). K can be parameterized using the data of a sonic anemometer, based

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on the friction velocity (u -star), the Von Karman-constant, the effective height and the stability factor (ζ) (Foken, 2006).

2.4 The flux chamber technique

Soil collars (50 cm × 50 cm) were inserted until 10 cm depth a week before the start of the experiment. Positions of soil collars were checked for being undisturbed and representative. The flux chambers (open dynamic chambers, 50 cm × 50 cm × 50 cm, produced by Karlsruhe Institute of Technology, Germany) consisted of a stainless steel frame, glass walls (transparency in UV (280–400 nm) ~ 50 %, transparency in visible light (400–700 nm) ~ 90 %), and a vent tube, and were tightened by use of clamps and rubber air strips. Two fans per flux chamber were continuously running, insuring well-mixed headspace air. Automatic chamber closure (once per hour) was made possible by use of a pneumatic system regulated by the valve manifold box. Air flow from the flux chambers to the FTIR was initiated by a membrane pump placed behind the measurement cell, set to 1 L min⁻¹. Air flow was measured every 2 min continuously for 20 min in flow mode. Chamber opening and closure was respectively after 4 and 18 min. Sampling lines from the chambers were of equal size and material and were tested for leaks regularly. Chamber temperatures were recorded by temperature loggers (Votcraft DL-1181THP). Fluxes were derived from concentration increases after chamber closure, by use of linear regression. Gas fluxes were calculated by:

$$F = \frac{VP}{RST} \frac{\delta C}{\delta t} \quad (2)$$

wherein V is the volume of the chamber (m³), P the chamber air pressure (Pa), R the gas constant (8.314 m³ Pa K⁻¹ mol⁻¹), S the chamber surface area (m²), T the chamber air temperature (K) and $\delta C/\delta t$ is the gas concentration change over time (mol mol⁻¹ s⁻¹). For flux calculations, only the concentration increases between 2 and 10 min after closure were used. Concentration increases were checked for non-linear

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trends and, if found, not used. Flux standard deviations were derived from the propagated standard deviations of the regression slope.

When homogeneity in footprint can be assured, micrometeorological and FC methods can be compared and used to study the role of photodegradation. Flux chambers can be shielded from incoming radiation, preventing photodegradation-induced carbon production, while micrometeorological methods capture all fluxes. Comparing the two methods therefore gives an indication of the presence and the magnitude of photodegradation-induced carbon fluxes (Rutledge et al., 2010). The use of this method was planned for our field experiment, but could not be applied due to lack of conformity between flux methods footprints; sparse active vegetation (with photosynthetic activity) was only present in the footprint of the FG technique, causing the methods to be incomparable.

To study photodegradation, two different flux chambers, one with and one without solar radiation exposure, were used. During this experiment, the flux chambers were measuring six fixed chamber locations, chambers were manually moved every few days. One flux chamber was made opaque by use of light excluding aluminium foil (on 5 August). On the days before (3–5 August), the positions were compared by measuring both locations with transparent chambers; both locations showed very similar CO₂ and CO flux patterns. Unfortunately, on 8 August, a leak was introduced in the opaque chamber system, wherefore direct comparison between the two treatments is limited to 3 days. With blank measurements, the flux chambers were tested for internal CO₂ and/or CO production. Minor CO production was found during the day, negligible in comparison to field CO production: values presented in this paper are not corrected for this.

Studying thermal degradation-induced CO₂ production in the field is not possible due to the simultaneous temperature response of biological CO₂ production. For CO, no temperature dependent biological CO production is expected, wherefore measurement of thermal degradation-induced CO production in the field is possible. To study the

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role of thermal degradation in field CO exchange, chamber temperature sensors were installed, measuring air temperature every minute.

2.5 Laboratory experiment

Two different laboratory experiments were performed to study photo- and thermal degradation. Grass samples for the laboratory experiment were taken from the field site (senescent above ground grass material) and mixed soil material samples were taken from the upper 3 cm of the soil. Both sample types were dried at 35 °C, to assure microbial activity to be negligible (Lee et al., 2012).

Photodegradation of senescent grass material was studied with a set up consisting of a metal cylinder with a plexiglass cap (transparent to UV) below an high intensity (above natural values) UV-A and UV-B source (UV-A lamp (Phillips TL 60W/10R, peak emission at 375 nm ($\pm 45 \text{ W m}^{-2} \text{ nm}^{-1}$), UV-B-lamp (Phillips TL 40W/12RS, peak emission at 310 nm ($\pm 30 \text{ W m}^{-2} \text{ nm}^{-1}$) Isitec GmbH, Bremerhaven). During the experiment, different samples (empty cilinder, 2 gram-sample and 4 gram-sample) were exposed to different amounts of radiation (no radiation, UV-A and/or UV-B radiation). During the experiments, air was continuously circulated from the cylinder to the FTIR and measured once per minute; emissions were derived from the measured concentration changes. Cylinder temperatures were monitored by an internal temperature probe (GTH 175/PT, Greisinger Electronics) and remained constant over the experiments ($21 \pm 0.3 \text{ }^\circ\text{C}$). Every experiment was performed twice.

To study thermal degradation, a glass flask was placed in a closed loop with the FTIR. For this experiment, only glass and stainless steel materials were used. Grass samples (4 samples of 2 grams) and soil samples (4 samples of 30 grams) were heated in temperature steps of 5° (20–65 °C) by use of a controlled temperature water bath. During the experiments, air was circulated from the glass flask to the FTIR and measured once per minute; emissions were derived from the measured concentration changes. Glass flask air temperatures were manually measured to check if water bath temperature was representative for grass and soil material temperatures; after 5 min, the glass

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flask air temperature had reached the same temperature as the water. All experiments were performed in duplicate and in dark conditions.

In the results sections, the given regression coefficients from polynomial fits are the explained sum of squares divided by the total sum of squares.

3 Results

3.1 Flux gradient measurements

During the field campaign (3 August–11 September, 2013), total precipitation was 15 mm and air temperatures ranged between 13 and 43 °C (see Fig. 2). Soil water content, measured at 10 cm depth was 18 % (VWC) and decreased less than 1 % over the experiment. FG measurements were done at the same location as EC measurements. During day time, footprint analysis showed that 90 % of the source area of the EC signal came from within 150 m, from within the grassland area. Since the FG method is measuring at the same location and height, it is expected that daytime FG fluxes mainly originate from the grassland area as well. During nighttime, footprint analysis showed fluxes mainly originating from outside the grassland. FG CO₂ fluxes ranged between -7 and 8 μmol m⁻² s⁻¹ (Fig. 1).

FG CO uptake (up to 1 nmol m⁻² s⁻¹) and emission (up to 2 nmol m⁻² s⁻¹) at night were observed. During the day, large (≥ 10 nmol m⁻² s⁻¹) CO emissions were visible (Fig. 1). Based on FG measurements, net ~ 42 (±30) nmol CO m⁻² per day is estimated to be emitted.

3.2 Flux chamber measurements

Air temperatures, FC CO₂ and FC CO fluxes can be seen in Fig. 2, rain events are indicated. FC CO₂ fluxes showed a diurnal pattern with small emissions at night (1 μmol m⁻² s⁻¹) and higher emissions during the day (up to 8 μmol m⁻² s⁻¹). Large rain

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events on 20 and 27 August (6.6 and 2 mm) caused a short increase in chamber CO₂ fluxes. Locations without organic surface material showed slightly lower CO₂ (and CO) fluxes.

At night, CO uptake of maximum 0.8 nmol m⁻² s⁻¹ was observed. During the day, emissions up to 3 nmol m⁻² s⁻¹ were observed. Over the course of the experiment, nightly CO uptake was continuously decreasing. The rain events caused a clear increase in nightly CO uptake, after which the decreasing continued (Fig. 2). Based on FC measurements, net ~ 8 (±1.2) nmol CO m⁻² per day is estimated to be emitted.

3.3 Photo- and thermal degradation

Photodegradation was studied by comparing opaque and transparent chamber measurements of three days (5–8 August) and by analysis of transparent FC data of a period in August (period with fixed location, stable weather conditions and no precipitation).

Possible photo- and/or thermal degradation-induced CH₄ fluxes are not shown or evaluated here: FG CH₄ fluxes were too small for dependency analysis and CH₄ chamber fluxes mostly showed uptake, indicating a different process than thermal or photodegradation.

3.3.1 CO₂ fluxes

Figure 3 shows the CO₂ fluxes (of transparent and opaque chamber) vs. air temperatures (Fig. 3a) and chamber temperatures (after 6 min closure, Fig. 3c). FC measurements showed very weak dependency on soil temperatures at 10 cm (not shown). Blocking radiation showed no distinguished impact on measured CO₂ fluxes. Chamber CO₂ fluxes correlate well with air temperatures and less with chamber temperatures (Fig. 3a and c). Chamber coverage had an effect on chamber temperatures; during daytime hours, the opaque chamber temperature differed up to 10 °C from the transparent chamber temperature.

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3.3.2 CO fluxes

A clear effect of chamber coverage on CO fluxes was visible; transparent chamber fluxes were significantly higher during the day. FC CO fluxes correlate better with chamber temperatures than with air temperatures (Fig. 3b and d).

Figure 4 shows CO fluxes in the transparent chamber vs. air temperatures (Fig. 4a), chamber temperatures (after 6 min closure, Fig. 4b) and amount of solar radiation (Fig. 4c) for a period in August. Again, CO fluxes relate best to chamber temperatures, and less to air temperatures and amount of incoming radiation (Fig. 4).

A temperature dependent biological CO uptake curve was fitted over chamber temperature data from (cold) night conditions (when abiotic fluxes are assumed to be minimal) and extrapolated to warmer temperatures. For biological CO uptake, a Q10-value from literature of 1.8 was chosen (Whalen and Reeburgh, 2001). An abiotic thermal degradation Q10-curve was fitted, also based on chamber temperature data, with a fitted Q10-value of 2.1. The sum of both processes agrees well the observed field CO emissions ($R^2 = 0.85$, Fig. 5).

3.3.3 Laboratory experiment

In the laboratory, exposure of senescent plant material from the field site to high intensity UV radiation did not result in increased CO₂ or CO fluxes in comparison to measurements performed in dark conditions.

Grass and soil material samples exposed to different temperatures, under dark conditions, showed significant CO₂ production during lower temperatures (< 40 °C) and displayed small CO₂ emissions at higher temperatures (> 55 °C) (Fig. 6a). For CO, clear thermal production was found, exponentially increasing with higher temperatures (Fig. 6b). A Q10-value of 2.14 (senescent grass material) and 2.00 (soil material) was found to fit best to the observed laboratory thermal degradation CO fluxes (Fig. 6b).

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4 Discussion

4.1 CO₂ fluxes

EC and FG measurements showed that the arid grassland was not yet in dormant state; significant CO₂ uptake was observed during the day. FC CO₂ measurements, performed on locations without active vegetation, solely showed positive CO₂ fluxes, with peak emissions during the day up to 8 μmol m⁻² s⁻¹. Figures 3a and 4a show that CO₂ fluxes mostly relate to air temperatures, and poorly relate to soil temperatures (not shown). Expected is that most CO₂ production takes place close to the surface where the temperature follows air temperatures closer than it follows soil temperatures at 10 cm depth. In the flux chambers, the rain events resulted in an increase in CO₂ production for several days, showing the typical water-dependent response of arid ecosystem respiration (Fig. 2).

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The simultaneous use of opaque and transparent chambers was employed to study the effect of radiation on carbon fluxes in the field. Blocking radiation had no visible effect on field chamber CO₂ fluxes (Fig. 3a and c). CO₂ flux measurements performed on bare soil locations seemed lower than other locations; senescent surface material seemed to contribute to total CO₂ fluxes (Fig. 2b). However, only 3 days of bare soil measurements are available and no opaque chamber measurements on bare soil are present, wherefore comparison is restricted.

In the thermal degradation laboratory experiment, CO₂ production from senescent plant and soil material was observed during lower temperatures (20–40 °C), indicating remaining biological activity, even after drying. Above 50 °C, an increasing CO₂ production was observed with increasing temperatures, therefore expected to be (partly) of non-biological origin. Possible abiotic CO₂ production of ±3 nmol min⁻¹ gr⁻¹ for senescent grass material was observed. Extrapolating the thermal production rates of

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the senescent grass material to field conditions (assuming 200 gr of senescent plant material per m^2 at 55°C), would result in a minor flux of $0.01 \mu\text{mol m}^{-2} \text{s}^{-1}$ (in comparison to observed field fluxes of $> 1 \mu\text{mol m}^{-2} \text{s}^{-1}$). Based on the observations in the laboratory, it is expected that the soil material also produces thermal degradation-induced CO_2 fluxes. However, considering the relative cold and wet conditions of the subsurface soil material in the field (compared to laboratory conditions and to surface temperatures), expected is that soil thermal degradation fluxes are minor in comparison to soil biological fluxes.

Other studies have observed thermal degradation-induced CO_2 fluxes with higher rates ($\pm 100 \text{ nmol CO}_2 \text{ gr}^{-1} \text{ min}^{-1}$ for C3-grass at 55°C), also at lower temperatures (Lee et al., 2012). We can not verify this observation for our field material. Based on our observations, we propose that under *natural* conditions, when soil surface temperatures and especially soil subsurface temperatures rarely exceed 55°C , thermal degradation-induced CO_2 fluxes do not play an important role in comparison to biological production, even in arid regions such as our study area.

We observed that chamber design can significantly influence chamber temperatures: during mid-day, the opaque and transparent chamber temperatures could differ up to 10°C . As observed in the laboratory experiment, *unnatural* high temperatures might lead to abiotic thermal CO_2 production. A research set up aiming at measuring photodegradation can unintentionally result in high surface temperature levels, which could lead to unrepresentative high abiotic CO_2 production estimates.

Similar as what has been found by Kirschbaum et al. (2011); Lambie et al. (2014); Uselman et al. (2011), we did not find photodegradation-induced CO_2 fluxes in the field as well as in the laboratory. This is in contrast to other photodegradation (field) studies, which have observed photodegradation fluxes (Lee et al., 2012; Rutledge et al., 2010; King et al., 2012). Potential explanations for this difference are: (a) the used field methodology in previous studies was not suitable for measuring (direct) abiotic degradation fluxes; (b) the role and significance of photodegradation differs per material and per field site or (c) studies might (partly) have misinterpreted thermal degra-

dation fluxes as photodegradation fluxes. However, as shown, the magnitude and the potential importance of thermal degradation-induced CO₂ fluxes in arid ecosystems are still unknown.

4.2 CO fluxes

During the measurement period, both CO uptake as emission has been observed by the FG method (patches of green active vegetation inside the footprint) as well as by the FC method (no active vegetation contributing to the fluxes) (Figs. 1 and 2). During the night, uptake up to 1 nmol m⁻² s⁻¹ was observed, which is most likely caused by microbial oxidation to CO₂ or CH₄ (Ingersoll et al., 1974; Conrad, 1996; Spratt and Hubbard, 1981; Yonemura et al., 2000; Whalen and Reeburgh, 2001; Bartholomew and Alexander, 1979; Bruhn et al., 2013). The CO uptake was decreasing over time but a rain event caused an enhanced uptake for some days. Soil biota being responsible for the CO uptake seems plausible since the effect of drought (decreasing uptake over time) and the effect of the rain (enhanced uptake) indicate a biological process. Nevertheless, with solely biological CO uptake taking place, one would expect higher uptake during warmer temperatures and no CO emission should be observed. Expected is a abiotic counteracting production process taking place simultaneously, “buffering” the CO uptake. For this reason, CO deposition velocities could not be calculated.

Photo- and thermal degradation

We propose that the observed CO emissions in the flux chambers are caused by thermal degradation.

FG measurements showed CO emissions during the day as well as during the night, indicating that CO is not (solely) produced by photodegradation (Fig. 1b). By means of opaque chamber measurements, significant lower CO fluxes (in comparison to transparent chamber measurements) were measured. However, as described before, FC temperatures were strongly affected by the blocking of solar radiation. Analysis of CO

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fluxes showed a strong correlation with FC temperatures, indicating that not the absence of radiation, but the indirect effect on temperature caused the significant lower CO emissions (Figs. 3 and 4).

FC CO fluxes were ranging between -1 and $2.5 \text{ nmol m}^{-2} \text{ s}^{-1}$ and only originated from soil or surface litter, since active vegetation was absent. Measured CO emissions are higher than reported for CO emissions from living plants and similar to values found for senescent plant material (Bruhn et al., 2013; Zepp et al., 1998; Schade et al., 1999; Derendorp et al., 2011; Lee et al., 2012). However, the measurements are a cumulative signal of uptake and emission and can therefore not be compared directly to other studies.

In the laboratory experiment, in where grass from the fieldsite was exposed to above natural intensity UV radiation, no photodegradation-induced CO fluxes were observed. However, significant thermal degradation-induced fluxes from the senescent field site grass and soil material were measured, visible already at low temperatures (20°C). At 50°C , a thermal CO production rate of senescent grass material of $0.13 \text{ nmol min}^{-1} \text{ gr}^{-1}$ was found. Extrapolating this observation to field conditions (assuming 200 grams of senescent plant material per m^2 at 50°C), would result in a flux of $\pm 0.4 \text{ nmol m}^{-2} \text{ s}^{-1}$, which is approximately 5 times lower than the measured (net) field CO fluxes. Extrapolating the thermally-induced CO production rate of the soil material to field conditions would result in an estimated production of $\pm 1 \text{ nmol m}^{-2} \text{ s}^{-1}$ by the upper 3 cm of the soil during a summer day. However, while this estimate indicates that abiotic thermal soil CO production indeed might play a major role, for accurate estimates for net soil CO uptake or emission, more information about biological CO uptake and about the soil profile is needed.

The observed chamber CO fluxes are suggested to be a cumulative signal of biological uptake (taking place in the soil) and abiotic thermal degradation (taking place in the soil and on the surface). The sum of both processes was fitted over chamber temperatures ($R^2 = 0.84$). For biological CO uptake, a Q10-value of 1.8 was used (based

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on literature values). To reproduce the observed field chamber CO fluxes, a higher Q10-value (of 2.1) for abiotic thermal soil CO production was fitted.

The laboratory measurements were used to experimentally determine the Q10-value of thermal degradation-induced CO fluxes. Q10-values of 2.14 and 2.00 were found which are similar to the fitted Q10-value for thermal degradation (Fig. 5).

The soil CO uptake process, taking place below the surface, is subject to buffered chamber temperatures, and therefore the chosen Q10-value might be an underestimation. Also, the biological soil uptake is not expected to follow the Q10-temperature response at higher temperatures ($> 35^{\circ}\text{C}$). Nevertheless, the difference in temperature response (as a consequence of different Q10-values or as a consequence of buffered temperatures) causes biological CO uptake to be dominant during colder (chamber) temperatures, and thermal degradation to be dominant during warmer (chamber) temperatures. During our field experiment, thermal degradation started to be dominant from approximately 25°C (chamber temperature) and followed an exponential curve with higher temperatures (Fig. 5).

Average net (uptake and emission) CO exchange estimates from FG and FC measurements differed significantly. The difference is most likely caused by several factors. In the FG footprint, relatively more (higher) dead vegetation was present since, for practical reasons, the chambers were placed over low dead vegetation. Also, the FG footprint contained active vegetation, which is another possible CO emitting source (Bruhn et al., 2013). Furthermore, the flux chambers block some incoming radiation (due to the frame and the glass, leading to lower exposure to radiation than in natural conditions) but also have shown to increase chamber air temperatures, with an unknown overall effect. Nevertheless, measurements show that the field site is a net source of CO during the summer months, affecting the atmospheric chemistry, at least at plant level, via OH^- depletion. More field measurements on (annual) CO exchange are needed to better understand the role of thermal degradation in CO (and CO_2) exchange in arid regions.

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5 Conclusions

In our field and laboratory experiment, direct photodegradation-induced CO₂ and CO fluxes have not been observed. Based on laboratory experiments, the production of thermal degradation-induced CO₂ is expected, but only significant under unnaturally high temperatures. In the laboratory, thermal degradation-induced CO fluxes were clearly observed, also at relatively low temperatures (20 °C). In the field, as well biological CO uptake as well as abiotic CO production was observed; abiotic CO production is assumed to be mainly a product of thermal degradation. The Q10-value of the CO producing thermal degradation process, as determined in the laboratory, agrees well with the fitted Q10-value for abiotic CO fluxes measured at the fieldsite.

Not all litter types are reported to be sensitive to photodegradation, which could explain why we did not measure photodegradation-induced fluxes. Also, we realize that in field conditions, partitioning thermal degradation from photodegradation is challenging. We therefore do not exclude the existence of photodegradation. However, in our field experiment in an arid ecosystem, we were not able to observe any direct photodegradation-induced carbon fluxes, showing that direct photodegradation does not play a major role in this arid ecosystem. Previous studies suggesting the occurrence of major photodegradation fluxes might possibly have neglected thermal degradation fluxes, which is an indirect effect of radiation. The potential importance of abiotic decomposition in the form of thermal degradation, especially for arid regions, should be considered and be studied in more detail.

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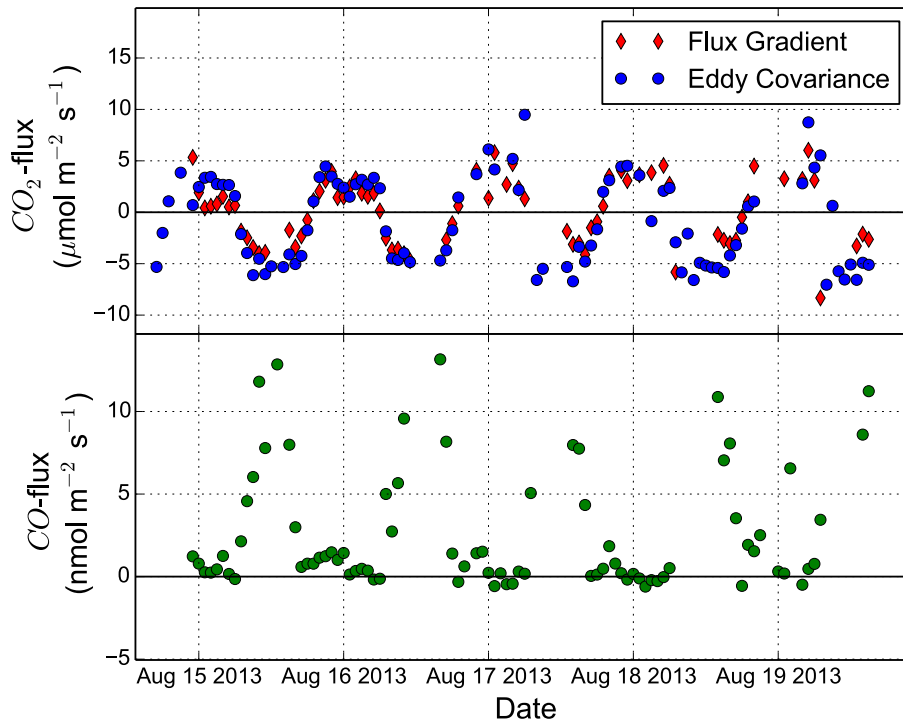


Figure 1. Five days of flux gradient- and Eddy Covariance- CO_2 and CO flux measurements.

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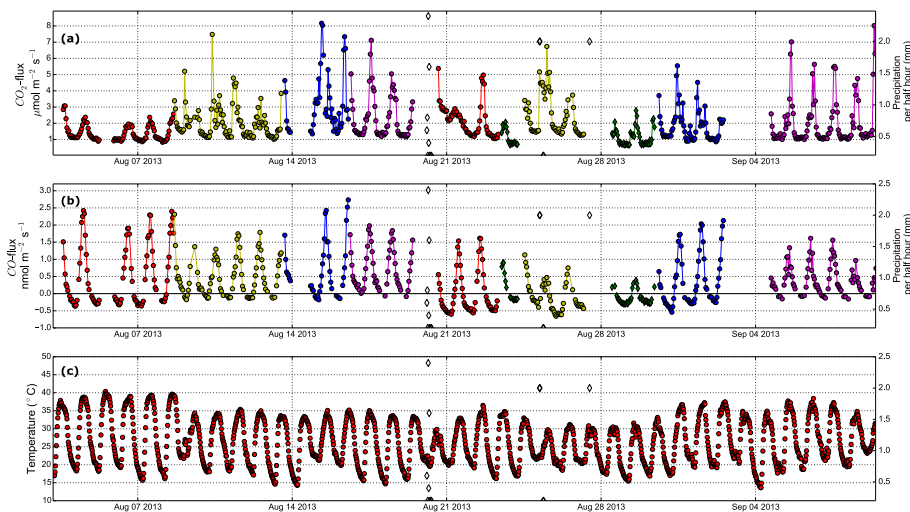


Figure 2. (a, b) Chamber CO_2 and CO fluxes with (errorbars with SD of flux are included but not visible due to low value) during field experiment, different colors are different locations, bare soil locations (green diamonds) and rain events (open diamonds) are indicated; (c) Air temperature ($^{\circ}\text{C}$).

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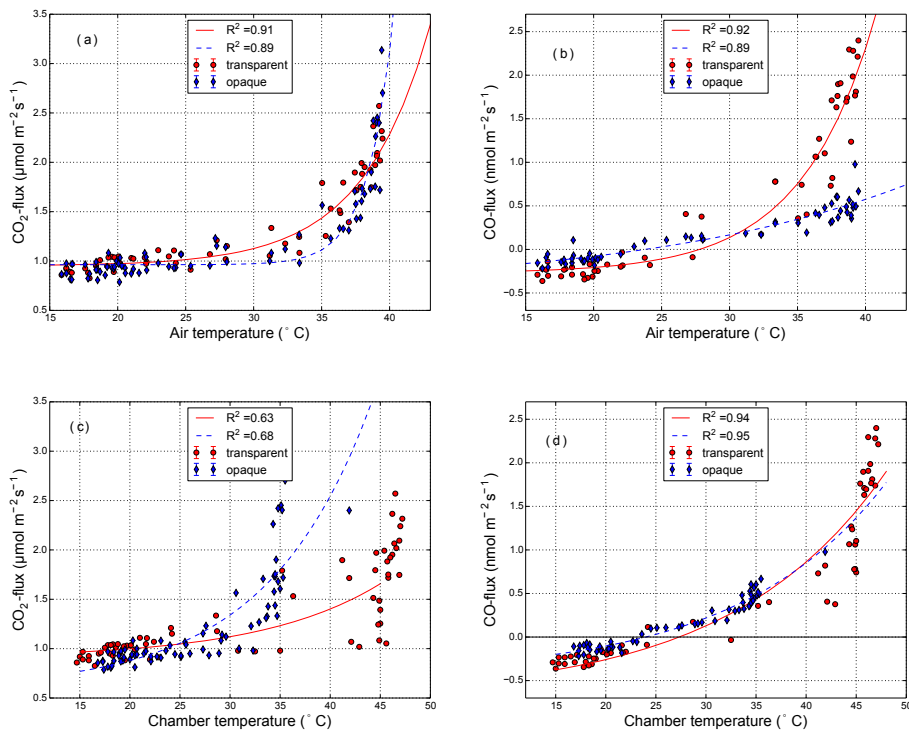


Figure 3. Chamber CO₂ fluxes (left) and CO fluxes (right) vs. air temperature (**a, c**) and chamber temperature after 6 min closure (**b, d**). Regression coefficients of polynomial fits are given in the legends.

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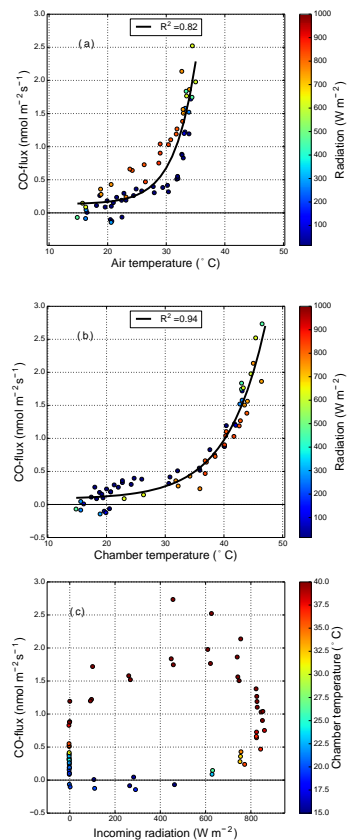


Figure 4. Transparent chamber CO fluxes for 15–19 August vs. air temperature **(a)**, chamber temperature after 6 min closure **(b)** and solar radiation **(c)**. Regression coefficients of polynomial fits are given in the legends.

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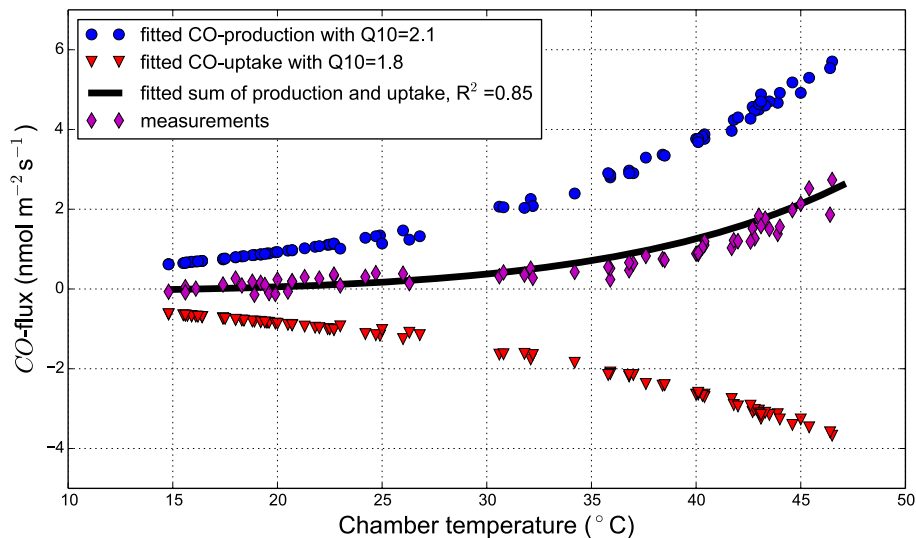


Figure 5. Fitted CO flux for 15–19 August, as a product of CO uptake ($Q_{10} = 1.8$, based on literature Whalen and Reeburgh, 2001) and CO production ($Q_{10} = 2.1$) based on chamber temperature (after 6 min closure) ($R^2 = 0.84$).

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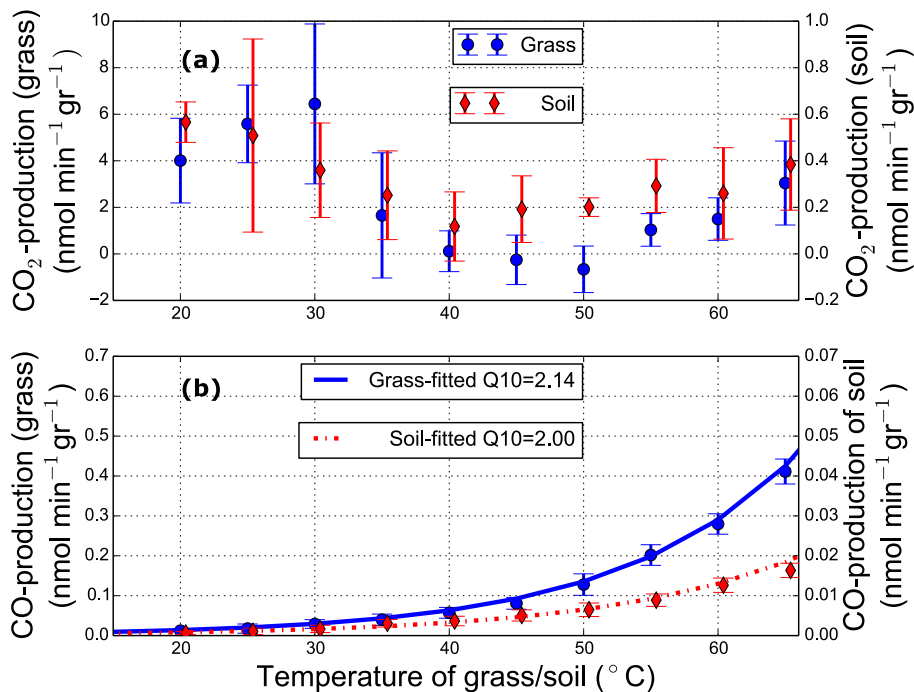


Figure 6. (a) Average CO₂ production of grass and soil material (nmol min⁻¹ gr⁻¹) over different temperatures in laboratory experiment; (b) Average CO production of grass and soil material (nmol min⁻¹ gr⁻¹) over different temperatures in laboratory experiment, with fitted Q10-value.