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Atmospheric $\Delta^{14}CO_2$ trend in Western European background air from 2000 to 2012

By INGEBORG LEVIN*, BERND KROMER and SAMUEL HAMMER,
Institut für Umweltphysik, Heidelberg University, INF 229, 69120 Heidelberg, Germany

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ABSTRACT

Long-term measurements of atmospheric $\Delta^{14}CO_2$ from two monitoring stations, one in the European Alps (Jungfraujoch, Switzerland) and the other in the Black Forest (Schauinsland, Germany), are presented. Both records show a steady decrease, changing from about 6‰ per year at the beginning of the century to only 3‰ per year on average in the last 4 yr. A significant seasonal variation of $\Delta^{14}CO_2$ is observed at both sites with maxima during late summer and minima in late winter/early spring. While the $\Delta^{14}C$ maxima are similar at Jungfraujoch and Schauinsland, the minima at Schauinsland are lower by up to 10‰, due to a larger influence from ^{14}C-free fossil fuel CO_2 emissions in the footprint of the Schauinsland station in winter. Summer mean $\Delta^{14}C$ values at Schauinsland are considered best suited as input for studies of biospheric carbon cycling in mid-northern latitudes or for dating of organic material of the last half century.

Keywords: carbon dioxide, radiocarbon, clean air reference

1. Introduction

The bomb radiocarbon signal in atmospheric carbon dioxide (CO_2) has been used as transient tracer in numerous applications, for example, to: (1) study the dynamics and transport processes in the atmosphere, hydrosphere and biosphere (e.g. Czeplak and Junge, 1974; Oeschger et al., 1975; Maier-Reimer and Hasselmann, 1987; Dörr and Münnich, 1986; Johnston, 1989; Trumbore, 2000; 2009); (2) constrain fluxes in the global carbon cycle (e.g. Siegenthaler et al., 1980; Randerson et al., 2002; Naegler, 2009; Naegler and Levin, 2009; Levin et al., 2010); and also (3) for dating of young organic material and in forensic studies (e.g. Wild et al., 2000; Spalding et al., 2005; Ubelaker et al., 2006). Common basis of these investigations are precise atmospheric $^{14}CO_2$ observations, which serve as an input or reference signal that is transferred into the carbon reservoir under investigation or that are used for dating at annual resolution. The radiocarbon measurement records available for Central European background stations, such as Vermunt, Austrian Alps; Schauinsland, Black Forest, Germany; and Jungfraujoch, Swiss Alps, covering the period from 1959 onwards (Levin and Kromer, 2004), have served as such a reference for applications in mid-latitudes of the Northern Hemisphere. Here, we present an extension of our measurements from the two stations Schauinsland and Jungfraujoch. These data are not only of importance for the above-mentioned applications, but Jungfraujoch measurements are also often used to define the European clean air reference when estimating regional fossil fuel CO_2 levels at polluted stations in Europe (e.g. Levin et al., 1980; Levin et al., 2003; Rakowski et al., 2004; Van Der Laan et al., 2010; Levin et al., 2011). In the following, we present and discuss the data covering the past decade (2000 to 2012). Earlier measurements are available in Levin and Kromer (2004), and in digital form under http://www.iup.uni-heidelberg.de/institut/forschung/groups/kk/Data_html. The new data presented here will be available at the same link.

2. Sampling sites and methods

The high-alpine monitoring station Jungfraujoch is located in the Swiss Alps (JFJ, Berner Oberland, 46°33′N, 7°59′E) at an elevation of 3450 m a.s.l. This high-elevation Global Atmosphere Watch (GAW) station is only occasionally – for example, during strong convective mixing conditions – influenced by regional CO_2 sources (e.g. Tucson et al., 2011), but for most of the time it samples air from the free troposphere over Europe

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*Corresponding author.
email: Ingeborg.Levin@iup.uni-heidelberg.de

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(Levin et al., 2008). The Schauinsland observatory in the Black Forest, situated on a mountain ridge at an elevation of 1205 m a.s.l. (SIL, 47°55′N, 7°54′E), is located at the eastern boarder of the upper Rhine valley and normally samples free tropospheric air during night. During the day and particularly in summer, Schauinsland station is frequently influenced by boundary layer air and moderate pollution events from the industrialised and populated Rhine valley (Schmidt et al., 2003). At both stations, JFJ and SIL, 2-week integrated CO_2 samples are collected by chemical absorption in carbonate-free concentrated sodium hydroxide solution (Levin et al., 1980). In the Heidelberg laboratory, CO_2 is extracted from the basic solution with phosphoric acid; samples are then purified over activated charcoal and measured by conventional counting (Kromer and Münnich, 1992). All $\Delta^{14}CO_2$ data presented here are reported as fractionation-corrected permil-deviations from Oxalic Acid standard activity corrected for decay (Stuiver and Polach, 1977); the measurement precision of individual samples is generally ±2‰ (1 sigma).

3. Results

The long-term decrease of Δ^{14}C in atmospheric CO_2 observed since the 1960s has continued in the last decade, albeit at a decreasing rate of only about 3‰ per year in 2007–2011. This decreasing trend reduces the precision of bomb ^{14}C dating compared to the preceding decades. The $\Delta^{14}CO_2$ decline today is driven primarily by the ongoing input of ^{14}C-free fossil fuel CO_2 into the global atmosphere, as the atmospheric bomb ^{14}C perturbation of the early 1960s has been almost fully equilibrated with surface ocean water and the terrestrial biosphere (Levin et al., 2010; Graven et al., 2012). Figure 1a shows individual Δ^{14}C data measured on the Jungfraujoch samples. In most years we observe a seasonal variation with minimum Δ^{14}C values in winter and spring and maxima in summer and autumn. The solid line in Fig. 1a is a fit curve calculated according to Nakazawa et al. (1997) through monthly mean Jungfraujoch data (Table 1). Levin et al. (2010) could show with the carbon cycle box model GRACE that this seasonality is mainly due to seasonal variations of the share of ^{14}C-elevated stratospheric air in the northern hemispheric troposphere and to a seasonally changing amount of fossil fuel CO_2 in the troposphere. At Schauinsland station (Fig. 1b), the seasonal variation of Δ^{14}C is larger by more than a factor of two compared to Jungfraujoch data. This is due to the closer proximity of the Schauinsland site to the fossil fuel sources, for example, in the Rhine valley. However, the summer values at Schauinsland are very close to those observed at Jungfraujoch, indicating that in summer European fossil fuel CO_2 emissions are smaller and diluted into a higher mixed layer depth than in winter (Levin et al., 2003).

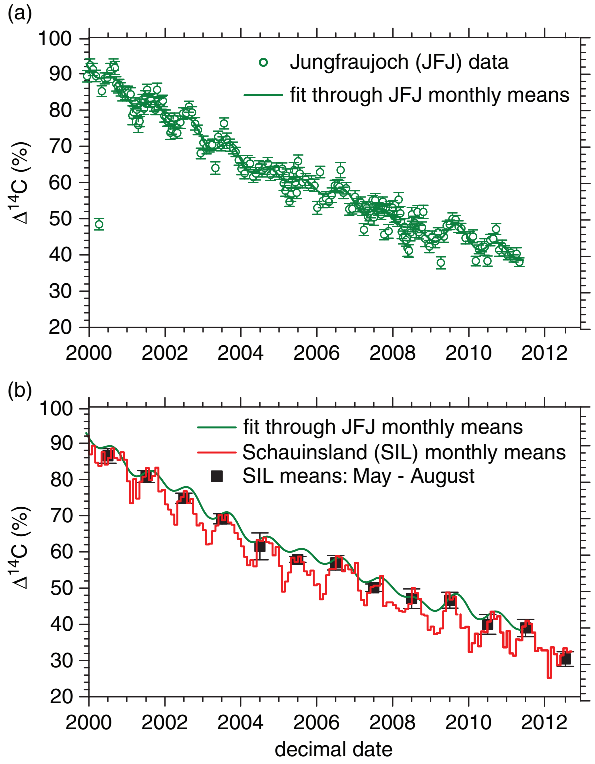

Fig. 1. $\Delta^{14}CO_2$ trends in background air over Europe 2000–2012. (a) Individual data from Jungfraujoch with 1σ error bars together with a harmonic fit curve calculated through the monthly mean data; the outlier in spring 2000 is a contaminated sample and is not included in the monthly mean values of Table 1, (b) Schauinsland monthly means in comparison with the Jungfraujoch fit curve from (a) and May–August (spring and summer) mean values with 1σ standard deviation of the four monthly values.

When searching for the proper European $^{14}CO_2$ reference curve for investigations linked to the terrestrial biosphere reservoir or for dating of recent organic material, the question is, which data set is most appropriate as input curve. Keeping in mind that photosynthesis by plants is mainly restricted to spring and summer months, and that generally the organic material to be dated has grown in rural areas and at lower elevation than Jungfraujoch, we suggest that only spring and summer months' data from SIL may best be taken for reference in such applications. Mean Δ^{14}C values for May to August have thus been calculated from monthly mean Schauinsland data and are also plotted in Fig. 1b, together with a 1σ standard deviation of the four monthly values. Our suggestion would be to use these numbers as input ^{14}C/C ratios which are transferred into plant material. However, there may be

Table 1. Monthly mean $\Delta^{14}CO_2$ data from Jungfraujoch and Schauinsland as well as monthly values from the fitted curve through Jungfraujoch data. Last column gives spring and summer mean values (including 1 sigma standard deviations of the monthly averaged data).

		Jungfraujoch	JFJ fit curve	Schauinsland	SIL summer
		Δ^{14}C	Δ^{14}C	Δ^{14}C	May–Aug
Decimal date	Month	[‰]	[‰]	[‰]	[‰]
2000.044	1	92.3	91.31	86.9	
2000.126	2	91.0	90.03	89.3	
2000.208	3	89.4	89.15	84.6	
2000.291	4	85.2	88.74	83.7	
2000.374	5	87.2	88.73	87.6	86.5 ± 2.0
2000.458	6	88.8	88.96	84.1	
2000.541	7	90.6	89.20	85.9	
2000.626	8	92.3	89.21	88.5	
2000.709	9	88.6	88.82	85.9	
2000.792	10	86.5	87.95	85.3	
2000.876	11	84.8	86.65	87.5	
2000.959	12	84.7	85.09	85.4	
2001.044	1	84.6	83.51	79.5	
2001.125	2	80.9	82.16	73.4	
2001.206	3	80.6	81.23	80.1	
2001.289	4	77.3	80.82	74.7	
2001.374	5	81.0	80.91	79.4	80.8 ± 1.6
2001.458	6	82.1	81.34	80.8	
2001.541	7	83.9	81.89	83.1	
2001.626	8	82.8	82.30	79.8	
2001.709	9	81.1	82.38	80.9	
2001.792	10	83.0	82.02	83.3	
2001.876	11	81.8	81.22	76.7	
2001.959	12	80.5	80.12	77.1	
2002.044	1	77.7	78.93	73.1	
2002.125	2	75.2	77.88	71.6	
2002.206	3	76.2	77.15	70.3	
2002.289	4	74.6	76.85	67.5	
2002.374	5	75.6	76.96	73.9	74.8 ± 1.4
2002.458	6	78.6	77.33	75.1	
2002.541	7	79.1	77.74	76.7	
2002.626	8	80.9	77.94	73.5	
2002.709	9	79.3	77.73	74.5	
2002.792	10	76.2	77.00	70.5	
2002.876	11	74.5	75.77	67.3	
2002.959	12	68.3	74.18	67.9	
2003.044	1	70.6	72.47	66.0	
2003.125	2	69.8	70.93	62.0	
2003.206	3	70.2	69.78	62.3	
2003.289	4	66.6	69.17	65.8	
2003.374	5	70.4	69.13	67.3	69.1 ± 1.4
2003.458	6	70.4	69.54	70.2	
2003.541	7	73.7	70.16	68.7	
2003.626	8	71.9	70.74	70.1	
2003.709	9	70.3	70.99	68.1	
2003.792	10	69.5	70.75	67.5	
2003.876	11	68.4	69.95	65.3	
2003.959	12	65.7	68.67	63.6	
2004.044	1	63.4	67.10	61.5	
2004.125	2	64.0	65.48	58.7	
2004.206	3	65.5	64.10	56.3	
2004.289	4	62.5	63.13	57.7	
2004.374	5	62.3	62.69	56.0	

Table 1 (*Continued*)

		Jungfraujoch	JFJ fit curve	Schauinsland	SIL summer
		$\Delta^{14}C$	$\Delta^{14}C$	$\Delta^{14}C$	May–Aug
Decimal date	Month	[‰]	[‰]	[‰]	[‰]
2004.458	6	64.3	62.74	63.4	61.5 ± 3.7
2004.541	7	63.3	63.13	62.9	
2004.626	8	64.0	63.66	63.9	
2004.709	9	65.2	64.10	62.6	
2004.792	10	62.1	64.05	62.3	
2004.876	11	63.1	63.47	59.0	
2004.959	12	63.4	62.62	61.4	
2005.044	1	63.1	61.69	56.1	
2005.125	2	60.1	60.86	49.0	
2005.206	3	60.1	60.28	51.8	
2005.289	4	56.1	60.03	54.3	
2005.374	5	62.3	60.09	58.4	57.9 ± 0.8
2005.458	6	59.3	60.36	57.2	
2005.541	7	63.9	60.66	58.7	
2005.626	8	60.1	60.82	57.1	
2005.709	9	60.8	60.71	54.6	
2005.792	10	59.3	60.28	53.6	
2005.876	11	60.0	59.55	54.8	
2005.959	12	57.2	58.65	50.8	
2006.044	1	58.0	57.76	51.1	
2006.125	2	58.0	57.06	46.9	
2006.206	3	55.4	56.68	48.4	
2006.289	4	54.8	56.69	53.6	
2006.374	5	56.4	57.03	55.3	57.0 ± 2.0
2006.458	6	59.2	57.57	55.2	
2006.541	7	59.1	58.11	58.8	
2006.626	8	62.0	58.43	58.5	
2006.709	9	57.1	58.38	54.6	
2006.792	10	57.8	57.88	56.3	
2006.876	11	52.6	56.95	55.2	
2006.959	12	54.0	55.71	54.1	
2007.044	1	54.7	54.37	56.9	
2007.125	2	54.2	53.14	49.3	
2007.206	3	50.5	52.21	46.8	
2007.289	4	52.9	51.70	45.3	
2007.373	5	51.0	51.62	48.7	50.1 ± 1.0
2007.456	6	53.0	51.88	50.3	
2007.540	7	53.2	52.31	50.7	
2007.625	8	52.8	52.71	50.7	
2007.708	9	52.9	52.89	53.3	
2007.792	10	49.6	52.74	45.0	
2007.875	11	49.7	52.22	46.1	
2007.959	12	52.4	51.40	46.1	
2008.044	1	51.3	50.40	45.4	
2008.125	2	51.9	49.40	44.4	
2008.206	3	49.3	48.56	43.7	
2008.289	4	46.2	47.97	43.5	
2008.374	5	42.5	47.65	44.2	47.1 ± 2.7
2008.458	6	46.0	47.56	45.6	
2008.540	7	49.1	47.56	50.0	
2008.625	8	49.3	47.50	48.6	
2008.708	9	46.6	47.26	47.0	
2008.792	10	49.1	46.77	42.7	
2008.875	11	44.5	46.06	42.4	

Table 1 (*Continued*)

Decimal date	Month	Jungfraujoch $\Delta^{14}C$ [‰]	JFJ fit curve $\Delta^{14}C$ [‰]	Schauinsland $\Delta^{14}C$ [‰]	SIL summer May–Aug [‰]
2008.959	12	42.3	45.22	38.4	
2009.044	1	45.0	44.43	39.4	
2009.125	2	43.8	43.87	37.3	
2009.206	3	45.3	43.70	37.6	
2009.289	4	39.4	43.98	38.9	
2009.374	5	46.3	44.71	43.6	46.7 ± 2.2
2009.458	6	48.2	45.74	48.8	
2009.540	7	49.5	46.85	46.8	
2009.625	8	49.8	47.80	47.6	
2009.708	9	47.8	48.36	42.7	
2009.792	10	47.3	48.38	na	
2009.875	11	44.7	47.80	39.0	
2009.959	12	44.7	46.73	39.3	
2010.044	1	45.1	45.33	32.3	
2010.125	2	45.1	43.88	34.8	
2010.206	3	38.8	42.61	37.4	
2010.289	4	41.5	41.73	33.9	
2010.374	5	42.2	41.36	38.0	40.0 ± 2.7
2010.458	6	39.9	41.48	37.8	
2010.541	7	42.6	41.97	40.7	
2010.626	8	44.5	42.63	43.5	
2010.709	9	47.1	43.22	41.5	
2010.792	10	41.4	43.47	42.2	
2010.876	11	42.5	42.97	37.7	
2010.959	12	41.1	42.13	33.6	
2011.044	1	39.7	41.10	38.2	
2011.125	2	40.5	40.08	32.0	
2011.206	3	39.3	39.23	31.4	
2011.289	4	39.5	38.69	35.8	
2011.374	5	38.0	38.49	35.6	39.0 ± 2.4
2011.458	6			39.4	
2011.541	7			41.0	
2011.626	8			40.1	
2011.709	9			37.8	
2011.792	10			32.6	
2011.876	11			33.4	
2011.959	12			32.8	
2012.044	1			32.8	
2012.125	2			25.2	
2012.206	3			33.7	
2012.289	4			31.8	
2012.374	5			28.7	31.5 ± 2.0
2012.458	6			31.4	
2012.541	7			33.4	
2012.626	8			32.3	
2012.709	9			32.5	

other applications where different data selection may be appropriate. For this purpose, monthly mean Schauinsland values, respective values from the Jungfraujoch together with the monthly data from the Jungfraujoch fit curve are listed along with the SIL summer mean values in Table 1. The individual 2-week integrated data are available online under http://www.iup.uni-heidelberg.de/institut/forschung/groups/kk/Data_html.

4. Summary and conclusions

Long-term integrated $\Delta^{14}CO_2$ measurements have been continued at the high-alpine Jungfraujoch station in the Swiss Alps as well as at Schauinsland in the Black Forest, southern Germany. The almost exponential decrease of $\Delta^{14}C$ since 1963 has continued with a current rate of ca. 3‰ per year in the last 4 yr. We suggest using the Jungfraujoch fit curve as a clean air reference for estimates of the fossil fuel CO_2 concentration at polluted European stations, while mean summer (May–August) data from Schauinsland may best represent atmospheric $\Delta^{14}C$ transferred into the biospheric reservoir. It should be noted, that the mid latitude northern hemispheric reference values presented here may not be applicable for tropical and southern hemispheric studies, as the increasing fossil fuel CO_2 emissions in northern mid-latitudes tend to increase the meridional gradient, so that respective background $\Delta^{14}C$ data may be higher in latitudes further to the south by as much as 5‰ (see Levin et al., 2010; Graven et al., 2012). Continuation of our long-term measurements seems appropriate as these data sets are essential as input to study carbon cycle dynamics or for future dating purposes.

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