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THE TROPOSPHERIC $^{14}CO_2$ LEVEL IN MID-LATITUDES OF THE NORTHERN HEMISPHERE (1959–2003)

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ABSTRACT. A comprehensive tropospheric $^{14}CO_2$ data set of quasi-continuous observations covering the time span from 1959 to 2003 is presented. Samples were collected at 3 European mountain sites at height levels of 1205 m (Schauinsland), 1800 m (Vermunt), and 3450 m asl (Jungfraujoch), and analyzed in the Heidelberg Radiocarbon Laboratory. The data set from Jungfraujoch (1986–2003) is considered to represent the free tropospheric background level at mid-latitudes of the Northern Hemisphere, as it compares well with recent (yet unpublished) measurements made at the marine baseline station Mace Head (west coast of Ireland). The Vermunt and Schauinsland records are significantly influenced by regional European fossil fuel CO_2 emissions. The respective $\Delta^{14}CO_2$ depletions, on an annual mean basis, are, however, only 5‰ less than at Jungfraujoch. Vermunt and Schauinsland both represent the mean continental European troposphere.

INTRODUCTION

The variations of radiocarbon in atmospheric carbon dioxide over the last 50 yr have been used in numerous studies of the global carbon cycle (e.g. Oeschger et al. 1975; Broecker et al. 1980; Siegenthaler 1983; Hesshaimer et al. 1994; Randerson et al. 2002) to determine the atmosphere-ocean CO_2 exchange rate (e.g. Wanninkhof 1992), soil carbon turnover (e.g. Dörr and Münnich 1989; Harrison et al. 1993; Trumbore et al. 1996), and regional fossil fuel CO_2 contributions (e.g. Tans et al. 1978; de Jong and Mook 1982; Levin et al. 1989; Levin et al. 2003). Moreover, present-day atmospheric $^{14}CO_2$ measurements are important for forensic investigations as well as for dating of post-bomb organic specimens. Systematic global observations of $^{14}CO_2$ in the troposphere were made during and after the atmospheric nuclear weapon tests in the 1950s and 1960s by several laboratories (e.g. Nydal and Lövseth 1983; Manning et al. 1990; Levin et al. 1985; Levin et al. 1992). Most observational programs were, however, terminated soon after the last atmospheric tests, when bomb $^{14}CO_2$ had been almost evenly distributed in the global troposphere.

As one of very few, the observational network of the Heidelberg Radiocarbon Laboratory has been maintained until today (Levin and Hesshaimer 2000). Although samples are collected at 3 sites in the Southern Hemisphere as well [and at 1 site in the tropics (Rozanski et al. 1995)], analyses are mainly restricted to the Northern Hemispheric (European) sites. Here, our primary application is to quantify fossil fuel CO_2 over western Europe (Levin et al. 2003). The aim of the present paper is to make our long-term $^{14}CO_2$ observations at mid-Northern Hemispheric sites available to the scientific community in order to serve as atmospheric input for global and regional carbon cycle investigations as well as for post-1950 dating applications. For these purposes, we have compiled time-weighted monthly mean values for Jungfraujoch (46°33′N, 7°42′E, 3450 m asl) and Schauinsland (47°55′N, 7°55′E, 1205 m asl, Levin and Kromer 1997) as well as composite annual means from both sites, in tabulated form. We also give revised annual mean $^{14}CO_2$ values for the Vermunt station (47°4′N, 9°34′E, 1800 m asl, 1959–1983) already published by Levin et al. (1985), but do not discuss this data set further in the present paper. The individual $^{14}CO_2$ analyses from all 3 sites are available as supplementary material (*Radiocarbon*, http://www.radiocarbon.org/IntCal04 and CDIAC, http://cdiac.esd.ornl.gov/trends/co2/cent.htm).

EXPERIMENTAL

Atmospheric $^{14}CO_2$ samples, integrated over bi-weekly intervals, have been collected from 1959 to 1986 at the Alpine station Vermunt, Austria; at the continental mountain station Schauinsland, Black Forest, Germany, since 1977; and at the High Alpine Research station Jungfraujoch, Switzerland, since 1986. $^{14}CO_2$ sampling and analysis techniques are described by Levin et al. (1980) and Schoch et al. (1980). $\delta^{13}C$-corrected $\Delta^{14}C$ data are given relative to NBS oxalic acid activity, corrected for decay (Stuiver and Polach 1977). Internal measurement precisions of individual samples typically are about $\Delta^{14}C$ = ±5–8‰ for Vermunt, about $\Delta^{14}C$ = ±3–5‰ for Schauinsland, and about $\Delta^{14}C$ = ±2–4‰ for Jungfraujoch. Samples from later years, and especially those from the clean air site Jungfraujoch, were measured to higher precision.

RESULTS

Figure 1a shows all (378) individual measurements performed at Jungfraujoch from July 1986 to July 2003 available to date. The smooth curve in Figure 1a presents a harmonic fit with a quadratic trend using the fitting routine of Nakazawa et al. (1997). Only 7 Jungfraujoch samples do not fall within a 3-σ range (1 σ = 2.8‰) around the harmonic fit curve. These outliers have been disregarded in the calculation of monthly and subsequently annual mean values for this site. There is a significant seasonal cycle observed in $\Delta^{14}CO_2$ at Jungfraujoch with peak-to-peak amplitudes between 5 and 8‰. Minimum values are observed in March and maximum values in August. This seasonal cycle is more pronounced in the first and the latest parts of the record than between 1992 and 1998. This seasonality is partly due to seasonal variations of stratosphere-troposphere exchange, and partly due to seasonal ^{14}C disequilibrium fluxes between the biosphere and the atmosphere (Hesshaimer 1997; Randerson et al. 2002). Monthly mean values of Jungfraujoch are plotted in Figure 1b together with the respective Schauinsland means. These values are given in Table 1. The Schauinsland $^{14}CO_2$ level is generally slightly lower than that at Jungfraujoch by about 2–6‰ in summer and about 10–15‰ in the winter half-year. This is due to the fact that the Schauinsland observatory can be influenced occasionally by Rhine valley pollutant sources, while the Jungfraujoch station is normally situated in the free troposphere, particularly during winter. A comparison of the Jungfraujoch data with recent measurements made during marine background conditions at the station Mace Head, situated at the west coast of Ireland (53°19′N, 9°53′W, 25 m asl), shows no significant difference, not even during summer (see next paragraph).

Also plotted in Figure 1b are the annual mean records from Jungfraujoch and Schauinsland as given in tabulated form in Table 2. The mean difference of annual means between Jungfraujoch and Schauinsland over the 17 yr of observations is 4.1 ± 0.4‰. This corresponds to an additional fossil fuel contribution at Schauinsland compared to Jungfraujoch of only 1.4 ppm (Levin et al. 2003). Compared to the marine background station at Izaña (28°18′N, 16°29′W, 2367 m asl), the Jungfraujoch annual means are lower by only 2.2 ± 0.5‰ (Levin and Hesshaimer 2000), and compared to the marine background level at Mace Head by only 0.7 ± 1.5‰. There is no seasonality observed in the difference between Jungfraujoch and Mace Head. We are thus confident that the Jungfraujoch data represent the $^{14}CO_2$ background situation in mid-northern latitudes (40–50°N) to better than ±1‰ during winter, but also in the summer half-year when vertical mixing over the continent is enhanced and Jungfraujoch may well be influenced by ground-level European emissions. For continental European (and probably also North American) studies—i.e. for dating of organic material which did not grow under real free tropospheric conditions—the Schauinsland (and Vermunt) summer means (May–August) may, however, be more suitable than Jungfraujoch. The Schauinsland values are lower on average by 2.5 ± 0.5‰ compared to the annual mean values at Jungfraujoch, and are also given in Table 2 together with summer means for Jungfraujoch.

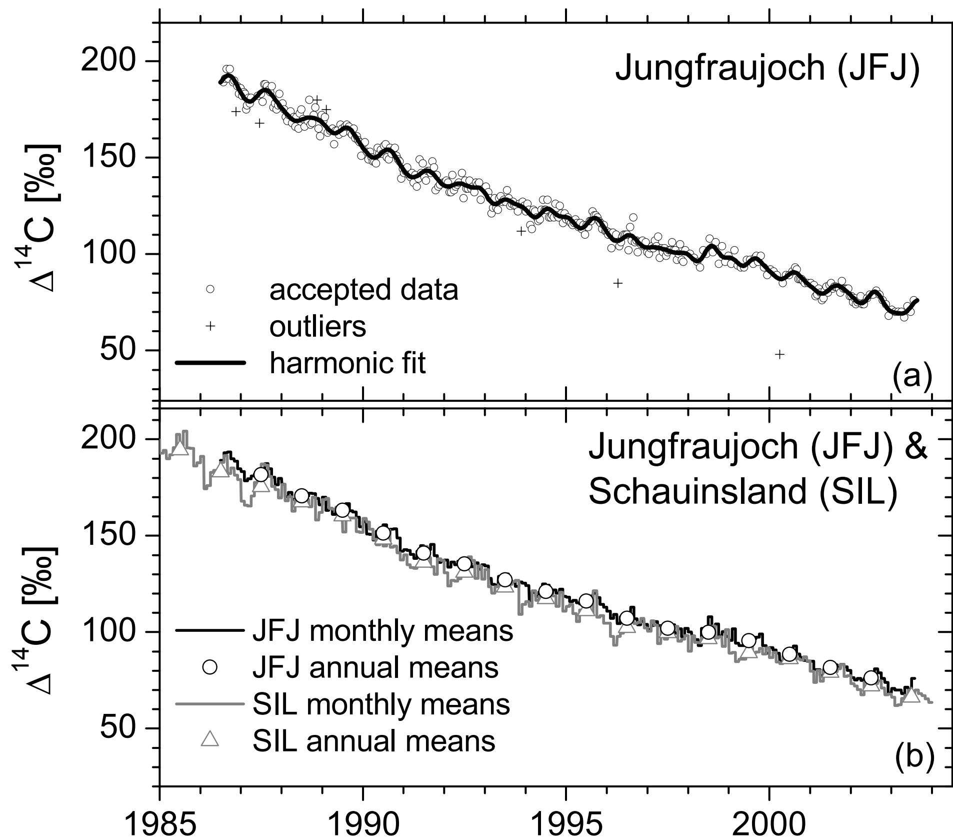

Figure 1 (a) Individual $\Delta^{14}C$ measurements determined on bi-weekly integrated atmospheric CO_2 samples from Jungfraujoch. The solid line is a harmonic fit curve calculated through the data. (b) Monthly and annual mean $\Delta^{14}CO_2$ values from Jungfraujoch and Schauinsland stations.

Annual mean values for the Vermunt site, already published by Levin et al. (1985), are here recalculated as time-weighted mean values from monthly data in the same way as for Jungfraujoch and Schauinsland, and we include these revised Vermunt data also in Table 2, as well as respective summer (May–August) means which extend the Schauinsland summer record back to 1959. The mean difference to our earlier Vermunt estimates is 0.1 ± 2.3‰.

CONCLUSIONS

Our combined precise data set from the 3 European sites Jungfraujoch, Vermunt, and Schauinsland provides the longest and most consistent atmospheric $^{14}CO_2$ record available to date. We hope that it is useful for various kinds of carbon cycle investigations and ^{14}C dating for samples grown after 1950. We are very much obliged to our teacher, the late Karl Otto Münnich, who, with wise foresight, pursued these measurements since almost the beginning of the nuclear weapon testing in the 1950s and always supported their continuation at very high quality.

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Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations.

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
1977	Jan	325.8	
1977	Feb	332.3	
1977	Mar	331.4	
1977	Apr	330.5	
1977	May	336.4	
1977	Jun	340.4	
1977	Jul	332.6	
1977	Aug	331.8	
1977	Sep	341.2	
1977	Oct	342.0	
1977	Nov	331.1	
1977	Dec	308.9	
1978	Jan	311.8	
1978	Feb	309.0	
1978	Mar	317.5	
1978	Apr	323.4	
1978	May	321.8	
1978	Jun	339.9	
1978	Jul	336.1	
1978	Aug	342.0	
1978	Sep	322.7	
1978	Oct	315.3	
1978	Nov	326.8	
1978	Dec	312.3	
1979	Jan	302.3	
1979	Feb	300.4	
1979	Mar	296.6	
1979	Apr	299.4	
1979	May	314.1	
1979	Jun	290.7	
1979	Jul	297.7	
1979	Aug	295.3	
1979	Sep	300.3	
1979	Oct	295.1	
1979	Nov	287.1	
1979	Dec	287.3	
1980	Jan	281.0	
1980	Feb	269.5	
1980	Mar	254.3	
1980	Apr	262.1	
1980	May	267.0	
1980	Jun	273.1	
1980	Jul	277.6	
1980	Aug	266.5	
1980	Sep	267.1	

Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations. *(Continued)*

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
1980	Oct	269.2	
1980	Nov	257.2	
1980	Dec	261.0	
1981	Jan	250.9	
1981	Feb	260.8	
1981	Mar	252.8	
1981	Apr	250.5	
1981	May	242.0	
1981	Jun		
1981	Jul	270.9	
1981	Aug	267.0	
1981	Sep		
1981	Oct		
1981	Nov		
1981	Dec		
1982	Jan	241.0	
1982	Feb	235.1	
1982	Mar	238.3	
1982	Apr	238.6	
1982	May	245.2	
1982	Jun	241.5	
1982	Jul	245.8	
1982	Aug	243.9	
1982	Sep	240.1	
1982	Oct	241.4	
1982	Nov	236.3	
1982	Dec	231.9	
1983	Jan	235.1	
1983	Feb	232.8	
1983	Mar	233.3	
1983	Apr	238.3	
1983	May	228.2	
1983	Jun	225.6	
1983	Jul	218.0	
1983	Aug	221.9	
1983	Sep	228.9	
1983	Oct	222.4	
1983	Nov	212.5	
1983	Dec	210.4	
1984	Jan	211.8	
1984	Feb	196.8	
1984	Mar	195.8	
1984	Apr	197.8	
1984	May	207.7	
1984	Jun	212.4	
1984	Jul	207.9	
1984	Aug	206.6	

Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations. *(Continued)*

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
1984	Sep	206.2	
1984	Oct	209.9	
1984	Nov	205.6	
1984	Dec	199.6	
1985	Jan	192.9	
1985	Feb	194.3	
1985	Mar	189.9	
1985	Apr	192.0	
1985	May	195.7	
1985	Jun	202.5	
1985	Jul	196.8	
1985	Aug	204.3	
1985	Sep	195.5	
1985	Oct	195.3	
1985	Nov	186.2	
1985	Dec	187.9	
1986	Jan	191.3	
1986	Feb	174.4	
1986	Mar	176.0	
1986	Apr	182.8	
1986	May	183.8	
1986	Jun	182.1	
1986	Jul	188.1	189.0
1986	Aug	191.6	193.0
1986	Sep	182.3	193.5
1986	Oct	185.2	189.9
1986	Nov	181.3	188.8
1986	Dec	179.2	184.8
1987	Jan	168.0	183.4
1987	Feb	165.9	178.5
1987	Mar	165.5	179.5
1987	Apr	170.6	181.0
1987	May	177.8	182.0
1987	Jun	182.3	182.5
1987	Jul	187.2	182.9
1987	Aug	186.6	187.5
1987	Sep	180.9	184.7
1987	Oct	175.6	180.4
1987	Nov	174.0	176.3
1987	Dec	169.6	179.7
1988	Jan	176.6	176.6
1988	Feb	168.1	170.2
1988	Mar	170.3	172.4
1988	Apr	162.8	169.0
1988	May	168.7	168.2
1988	Jun	169.5	172.3

Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations. *(Continued)*

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
1988	Jul	170.6	168.8
1988	Aug	168.7	170.6
1988	Sep	165.0	172.1
1988	Oct	169.9	171.8
1988	Nov	158.1	169.4
1988	Dec	162.1	166.0
1989	Jan	168.7	170.1
1989	Feb	169.3	163.1
1989	Mar	164.1	164.2
1989	Apr	163.9	161.0
1989	May	164.7	163.4
1989	Jun	159.8	165.5
1989	Jul	152.0	164.6
1989	Aug	158.4	166.7
1989	Sep	159.1	162.9
1989	Oct	159.3	162.4
1989	Nov	152.2	159.7
1989	Dec	151.7	154.6
1990	Jan	159.0	157.3
1990	Feb	157.1	151.0
1990	Mar	153.3	150.7
1990	Apr	148.3	147.7
1990	May	145.3	152.3
1990	Jun	151.6	152.9
1990	Jul	144.6	155.7
1990	Aug	152.9	150.3
1990	Sep	144.0	153.4
1990	Oct	144.6	153.5
1990	Nov	141.2	148.9
1990	Dec	137.4	142.0
1991	Jan	140.2	142.8
1991	Feb	135.0	142.4
1991	Mar	133.6	140.3
1991	Apr	133.1	138.3
1991	May	134.6	142.1
1991	Jun	135.8	144.8
1991	Jul	144.5	139.4
1991	Aug	138.7	142.8
1991	Sep	133.9	145.6
1991	Oct	130.0	139.8
1991	Nov	138.1	136.1
1991	Dec	137.1	136.9
1992	Jan	130.9	137.5
1992	Feb	123.8	133.6
1992	Mar	126.7	136.3
1992	Apr	126.5	135.0
1992	May	127.7	136.7

Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations. *(Continued)*

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
1992	Jun	134.4	135.6
1992	Jul	136.4	135.6
1992	Aug	139.1	135.8
1992	Sep	137.2	133.8
1992	Oct	127.8	135.9
1992	Nov	134.3	132.7
1992	Dec	127.5	135.0
1993	Jan	132.5	134.4
1993	Feb	123.6	127.9
1993	Mar	120.3	124.8
1993	Apr	123.5	124.3
1993	May	124.7	129.0
1993	Jun	125.4	127.5
1993	Jul	128.6	130.1
1993	Aug	124.7	126.4
1993	Sep	126.3	126.1
1993	Oct	126.9	123.7
1993	Nov	109.2	126.0
1993	Dec	114.4	125.4
1994	Jan	115.8	124.1
1994	Feb	113.3	116.3
1994	Mar	121.4	120.6
1994	Apr	116.4	118.5
1994	May	118.6	120.7
1994	Jun	120.4	125.1
1994	Jul	119.7	124.4
1994	Aug	119.4	119.3
1994	Sep	118.8	121.9
1994	Oct	113.3	120.1
1994	Nov	120.5	122.3
1994	Dec	110.9	118.6
1995	Jan	113.3	118.2
1995	Feb	114.3	115.9
1995	Mar	109.7	116.6
1995	Apr	105.5	115.3
1995	May	111.6	114.5
1995	Jun	110.7	112.7
1995	Jul	116.1	115.0
1995	Aug	109.4	118.7
1995	Sep	121.4	120.2
1995	Oct	107.7	118.0
1995	Nov	108.6	114.5
1995	Dec	104.8	113.0
1996	Jan	104.6	110.8
1996	Feb	97.7	107.1
1996	Mar	93.2	109.2
1996	Apr	97.8	104.4

Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations. *(Continued)*

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
1996	May	99.5	108.0
1996	Jun	104.7	110.0
1996	Jul	107.5	107.6
1996	Aug	105.7	112.9
1996	Sep	104.4	107.0
1996	Oct	105.1	103.7
1996	Nov	107.0	105.7
1996	Dec	102.3	104.1
1997	Jan	100.5	101.3
1997	Feb	105.9	103.5
1997	Mar	103.6	105.4
1997	Apr	94.0	103.3
1997	May	103.1	102.1
1997	Jun	96.4	100.6
1997	Jul	99.2	101.8
1997	Aug	101.2	103.8
1997	Sep	103.0	99.6
1997	Oct	95.3	101.8
1997	Nov	96.4	97.6
1997	Dec	98.3	102.0
1998	Jan	95.8	100.0
1998	Feb	97.9	98.0
1998	Mar	94.7	96.2
1998	Apr	94.5	93.0
1998	May	97.1	102.0
1998	Jun	97.3	100.0
1998	Jul	94.7	108.0
1998	Aug	103.3	101.2
1998	Sep	96.9	104.2
1998	Oct	97.6	98.0
1998	Nov	92.6	100.2
1998	Dec	101.3	97.9
1999	Jan	89.0	95.8
1999	Feb	89.4	101.8
1999	Mar	84.7	94.2
1999	Apr	86.0	93.0
1999	May	85.9	93.0
1999	Jun	91.0	97.0
1999	Jul	93.9	97.0
1999	Aug	89.3	96.6
1999	Sep	89.7	97.4
1999	Oct	91.5	99.0
1999	Nov	88.7	93.0
1999	Dec	91.8	89.0
2000	Jan	86.8	92.0
2000	Feb	89.4	90.8
2000	Mar	84.6	89.0

Table 1 Monthly mean $\Delta^{14}CO_2$ values from Schauinsland and Jungfraujoch stations. *(Continued)*

Year	Month	Schauinsland $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)
2000	Apr	83.6	85.0
2000	May	87.6	87.3
2000	Jun	84.3	89.0
2000	Jul	85.9	90.7
2000	Aug	88.6	92.8
2000	Sep	86.1	88.7
2000	Oct	85.6	86.7
2000	Nov	87.3	85.3
2000	Dec	85.2	85.0
2001	Jan	79.7	84.8
2001	Feb	73.9	80.5
2001	Mar	80.2	80.5
2001	Apr	74.6	77.4
2001	May	79.5	80.9
2001	Jun	80.8	82.0
2001	Jul	83.0	83.5
2001	Aug	79.5	83.0
2001	Sep	81.1	81.4
2001	Oct	83.3	83.3
2001	Nov	77.0	82.0
2001	Dec	77.4	80.1
2002	Jan	73.4	76.5
2002	Feb	72.0	75.2
2002	Mar	70.2	76.1
2002	Apr	67.6	75.0
2002	May	74.1	76.0
2002	Jun	75.3	78.7
2002	Jul	77.0	79.0
2002	Aug	73.2	81.0
2002	Sep	74.7	79.0
2002	Oct	71.0	76.0
2002	Nov	67.4	74.0
2002	Dec	67.8	68.3
2003	Jan	66.3	70.7
2003	Feb	62.0	70.0
2003	Mar	62.4	70.0
2003	Apr	65.7	68.2
2003	May	67.3	71.4
2003	Jun	70.0	70.3
2003	Jul	68.4	76.0
2003	Aug	70.0	
2003	Sep	68.1	
2003	Oct	67.2	
2003	Nov	65.3	
2003	Dec	63.6	

Table 2 Annual mean $\Delta^{14}CO_2$ values from Vermunt, Schauinsland, and Jungfraujoch stations as well as summer mean values for Schauinsland and Vermunt.

Year	Vermunt $^{14}CO_2$ (‰)	Vermunt (summer) $^{14}CO_2$ (‰)	Schauinsland $^{14}CO_2$ (‰)	Schauinsland (summer) $^{14}CO_2$ (‰)	Jungfraujoch $^{14}CO_2$ (‰)	Jungfraujoch (summer) $^{14}CO_2$ (‰)
1959	228.3	256.5				
1960	212.6	226.3				
1961	221.3	232.3				
1962	361.2	388.6				
1963	713.1	823.0				
1964	835.3	899.8				
1965	754.5	779.5				
1966	691.6	714.5				
1967	623.3	637.1				
1968	565.3	568.7				
1969	545.3	547.5				
1970	529.6	534.0				
1971	498.7	511.3				
1972	465.5	469.3				
1973	420.3	413.0				
1974	not enough data	402.5				
1975	not enough data	not enough data				
1976	350.5	349.8				
1977	333.6	337.6	332.0	335.3		
1978	324.3	327.0	323.2	334.9		
1979	293.7	295.3	297.2	299.4		
1980	263.8	266.5	267.1	271.1		
1981	256.3	262.7	256.4	260.0		
1982	238.8	238.7	239.9	244.1		
1983	226.6	228.7	225.6	223.4		
1984	not enough data	213.3	204.9	208.7		
1985	not enough data	206.4	194.4	199.8		
1986		190	183.2	186.4	not enough data	191.0
1987			175.3	183.5	181.5	183.7
1988			167.5	169.4	170.6	170.0
1989			160.3	158.7	163.2	165.0
1990			148.3	148.6	151.3	152.8
1991			136.2	138.4	140.9	142.3
1992			131.0	134.4	135.3	135.9
1993			123.3	125.9	127.1	128.2
1994			117.4	119.5	121.0	122.4
1995			111.1	111.9	116.0	115.2
1996			102.5	104.3	107.1	109.6
1997			99.7	99.9	101.9	102.1
1998			97.0	98.1	99.9	102.8
1999			89.2	90.0	95.6	95.9
2000			86.2	86.6	88.5	89.9
2001			79.2	80.7	81.7	82.3
2002			72.0	74.9	76.2	78.7
2003			66.4	68.9	not enough data	70.9