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Reconstruction of Atmospheric CO_2 Background Levels since 1826 from Direct Measurements near Ground

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Abstract

A new data set of annually averaged CO_2 background levels directly measured from 1826 to 1960 is presented. It is based on a selection process of about 100,000 single samples from more than 200,000 available near ground on land and sea, mainly in the northern hemisphere. Analysing the data, methods, sampling stations, meteorological conditions and air masses it is possible to reconstruct the past yearly CO_2 background levels. New methods to estimate annual marien boundary levels from near ground data from the historical data are presented. This allows the reconstruction within an estimated error range of ±2.5 % and a methodical error range since 1870 of ±3 %. A definite fluctuation of levels can be seen around 1860 and especially around 1940 showing levels of more than 380 ppm, almost like today. A slow rise of atmospheric CO_2 since 1880 is confirmed. The difference in averages of CO_2-levels in the 19th and 20th century of 2.6 % is within error range of 3 % of methods.

Keywords: CO_2 background level, marine boundry level, direct chemical methods, vertical profiles, wind and precipitation corrections.
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A summary of acronyms see at the end of the paper.

1. Introduction:

Atmospheric CO_2 concentrations have been measured since the beginning of the 19th century with remarkable precision. Carl Wilhelm Scheele was the first person, who from a number of ingeniously contrived experiments, concluded that the atmospheric air is a mixed fluid composed of about two parts of nitrogen, and one part of oxygen, along with a very small admixture of carbon dioxide. This statement by Joseph Black (1806), the discoverer of $CO_{2,}$ dates back to 1806 and marked the beginning of accurate gas analysis and the end of the phlogiston theory–a former heresy in science. (Rörsch 2007).

As outlined in Beck (2007, 2008) about 200 000 single samples of air have been collected and analysed by the scientists who established knowledge of modern natural science. Theodore de Saussure (1830)was the first who continuously analysed air samples in the Swiss alpine area since about 1809 up to 1830 and established accurate gas analysis by dissolving defined volumes of air in alkaline solution and subsequently dried and weighted them to calculate the corresponding concentration of CO_2 in air. He found the today well known characteristics of the atmospheric fluctuations of CO_2 during daytime and night time, the seasons, influence by vegetation and weather (fog, rain, snow) and burning fuels by man. Max Pettenkofer introduced the titrimetric CO_2 method around 1855 which became the standard in CO_2 gas analysis for the subsequent 100 years. Since about 1870 it was possible to validate the sampling and analyzing within an error range of ±3 % or about ±10 ppm.

In the 20th century hundreds of thousands of samples have been analysed on continents and over the oceans as well as investigating the vertical profile of air using a refined Pettenkofer method down to 0.33 % error in 1935. Modern NDIR spectroscopic analysis of air streams was introduced in 1938, exhibiting a much better accuracy of 0.1 % since the early 1950s and is still used for today´s atmospheric monitoring by NOAA. Meanwhile globally acting networks monitor CO_2 contents in the atmosphere or the hydrosphere as documented by NOAA GlobalView-CO_2 and WDCGG.

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†Post mortem memorial edition

In 1958 Keeling introduced continuous measurements of atmospheric CO$_2$ using modern NDIR procedure and defined the modern standards of background levels.

Historical CO$_2$ measurements prior to this year are usually derived from proxies, with ice cores being the favorite. Those done by chemical methods prior to 1960 are often rejected as being inadequate due too poor siting, timing or method. As outlined in Beck (2007) the vast majority of

these data has not been evaluated.

In this paper a new data set of annually averaged CO$_2$ background levels from 1826 to 1960 is presented based on the comprehensive analysis of direct near ground measurements. This was possible by the development of new methods to estimate the annual CO$_2$ background levels from near-ground data, as described in Massen et al. (2007).

2. Data and methods

An amount of more than 200,000 single samples is available in about 400 historical papers from 1800 to 1960 (Letts and Blake 1899–1902, Stepanova 1952).

The selection process leading from the raw data to the annual CO$_2$ background averages was as follows (Beck 2007, 2008 and Messen and Beck 2011):

1. Compiling data from available literature (data, stations, methods and literature in separate files)

   About 100 000 single samples (97,404 samples from 901 stations at sampling locations from lat -78.8 to +83.7 and long -156.8 to -163.3 most frequent locations: northern hemisphere at latitudes 40 to 80N (~75 %), Europe, averages of double and quadruple sampling) have been selected to calculate a dataset of monthly averages. There are 1172 months with data from 1800 to 1959 including 1998 with data sampled over the oceans, by a total 77 authors. Data from marine, coastal stations or from higher troposphere in 20th century are about 50 % of all. The longest series have been analysed at Montsouris (Paris) 1877–1910 (33 years).

   From 1949 to 1951 there has been measurements in a continuous air stream (URAS) as with the modern NDIR. Typical sampling height was 1–50 m above ground (sea surface), literature research has revealed that Andrée 1893, Wigand 1911, Kauko 1936, Bischof 1960 (Lapape 1928, 1929, 1934 ignored because of inconsistent data) have measured CO$_2$-levels at different altitudes (balloon, aeroplane) so that vertical profiles can be calculated.

   The most accurate analysis in 19th century had been conducted by Spring 1883 and Palmqvist 1889 (±1–2 %) using high precision and well calibrated chemical gas analysers. In the 20th century Kauko 1935 analysed vertical CO$_2$ profiles over Helsinki down to ±0.33 %, Kreutz analysed during 1939 to 1941 with ±1.5 % accuracy at 4 altitudes, 8 samples per day. An overall accuracy range of ±3 % can be recognized since 1870.

   The year 1960 marked the transition time when a methodical change occurred by no longer using old wet chemical methods, but new physical infrared spectroscopic NDIR methods by analysing a continous gas stream. Charles Keeling introduced the new method at the Mauna Loa Observatory (Hawaii) in 1958. It is remarkable that no calibration of the new NDIR method versus the old chemical methods can be found in literature.

   The data can be traced in two main libraries. 292 papers with CO$_2$ data in the 19th century are listed in Letts and Blake (1899). 231 papers with data from the 19th and 20th century have been compiled by Stepanova (1952). Additionally, the author has used 16 further papers showing CO$_2$ data not listed in the two libraries.

   Detailed station, sampling, author, and literature information are documented in the supplemental files to this paper.

2. Testing the reliability of data by checking the used methods, local sources and sinks, meteorological conditions (air mass analysis) and comparisons with other geophysical time series if possible.

3. Calculating monthly and yearly averages and estimating the annual background averages by using methods outlined in this paper. Often several data series per year are available. In total 10 linear interpolations fill the gaps of years with no data available since 1870 (31 since 1826)

   The selected dataset of about 90 000 values show very high values from 1800, analysed by de Saussure (1830).

   The very detailed information on methods, locations, and air masses at the time of the largest volcanic event in recent history (Tambora 1815–1816) justifies the implementation of his data. During expeditions to the Arctic circle and Antarctic areas, either very low values were sampled (Venus 1882 and Charcot 1908 expeditions, analysed by Müntz; Müller 1928, Buch 1936) or very high levels had been documented by Krogh 1902 and Lockhart 1941. The evaluation of the sampling procedures reveals accurate methods using calibrated gas analyzers by skilled persons. Therefore we have to accept these data as real such as high levels of CO_2 near the ice-covered waters. This was documented by different persons using different analysing methods. The very high levels seem to reflect known upwelling of CO_2-enriched waters.

   Some corrections have been applied to the dataset because of methodical errors existing in some French data series showing the sulphuric acid absorption problem, (Beck 2007). The Reiset, Müntz and Montsouris data (prior to 1890) have been corrected by an addition of about 20 ppm (20–30 ppm losses was demonstrated by Spring in 1883 using this procedure). Reiset passed his air samples through sulphuric acid, Müntz used a volumetric method and sulphuric acid to free the CO_2 from the alkaline carbonate. Data analysed at Montsouris can be confirmed because parallel analyses were done by other investigators, when the method was changed in 1890. Also the Brown and Escombe and Petermann (1892–93) data produced by a variant of the Reiset method (1898–1901) are influenced by using sulphuric acid absorption. Therefore a correction was necessary. The Feldt, Heimann and Frey data had to be corrected because of too short absorption time in alkaline solution. (For detailed informations please see in the station and author supplemental file)

Figure 1 shows a 3D presentation of the monthly averages according to latitudes. Figure 2a shows their sampling locations.

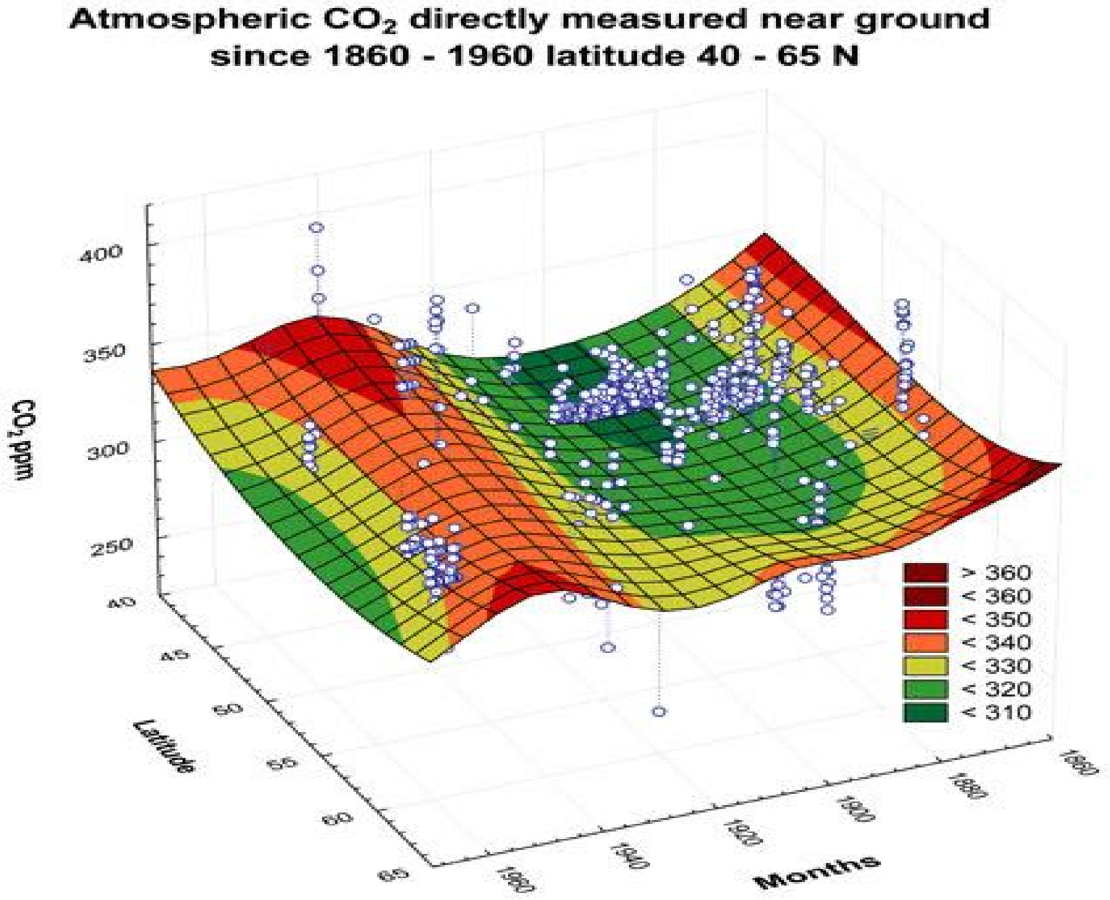


Figure 1. Monthly averages of about 72 % of raw CO_2 samples (latitude 40–65 N) collected 1860–1960 near ground and selected from >200 000 available in literature. Coloured plane = distance weighted least squares fit Statistica (2009); blue circles: monthly averages.

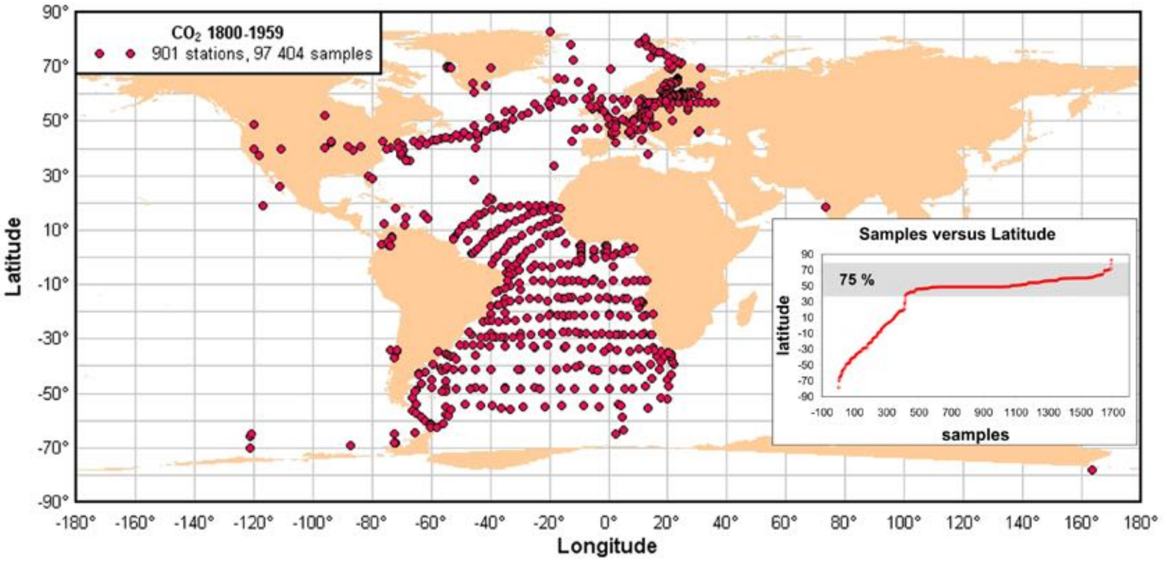
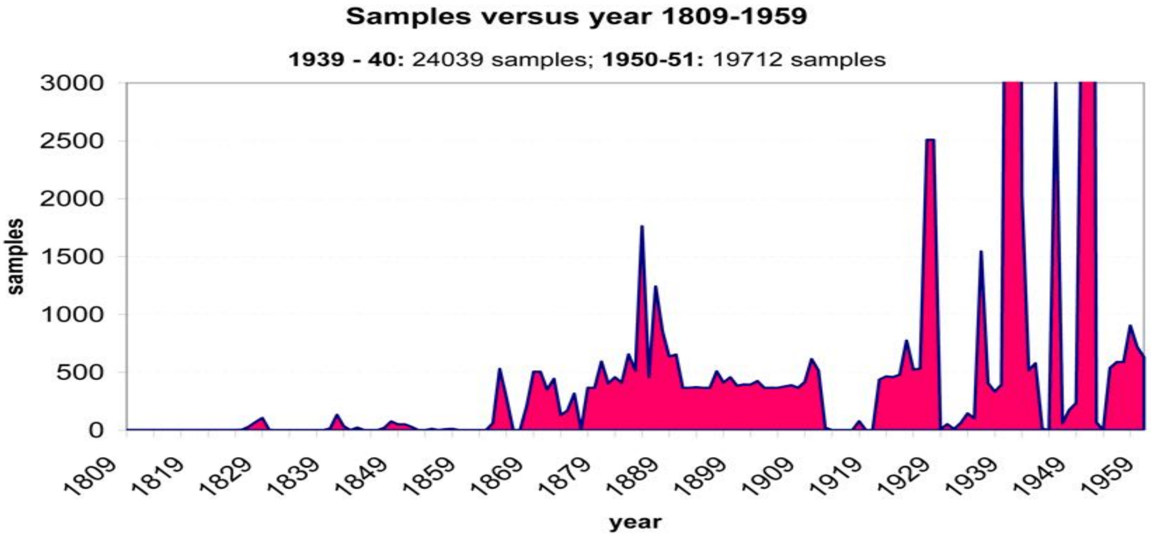


Figure 2a. 901 sampling stations of direct measured CO_2 near ground since 1800–1960 (dPlot2009). To the right: Samples versus latitude; 1172 months with data +Wattenberg/Buch single samples over sea. (e.g. modern WDCGG stations with monthly CO_2 data: 55, Globalview-CO_2 (2008):110)



Figur 2b. Samples per year (see supplemental file samplesused.pdf)

About 75 % of the samples were collected between latitudes of 40 and 80 N, about 50 % over sea surface or from the sea at coasts. Samples collected near the sea ice at the Arctic and Antarctic circle usually show very low values because of absorption in ice/water. Samples collected over sea surface near warm ocean currents or in upwelling areas show high values.

2.1 Summary of historical methods of gas analysis prior to ndir 1958

Since its early arising in the 18th century, chemical methods as described in Beck (2007, 2008) have been used to analyse fluid and gas mixtures. The typical procedure was to pass a defined volume of air through an alkaline solution of known concentration ($Ba(OH)_2$, NaOH, KOH) and calculate quantitatively absorbed CO_2 from the induced changes of the system. Up to about 1845 the gravimetric method was in favour by drying and weighting the produced carbonate. The French chemist Regnault introduced the volumetric method by measuring the changed volume of the system. No inter-calibration of the methods can be found in literature.

A landmark in gas analysis was established by the German physician Max v. Pettenkofer in 1857 when he developed a titrimetric method named in honour of him the “Pettenkofer Process”. It was based on the absorption of the CO_2 from air in baryta water and titration of the produced barium carbonate with acid. Since about 1855 inter-calibration of the several methodical variants are available. A typical analysis of an air sample lasted from several hours up to one day.

A theoretical accuracy of ±0.0006 % in volume was provided by the Pettenkofer process but in practice the accuracy was about ± 2 %, (Kauko et al. 1935). Special modifications of H. Lundegårdh (1922) resulted in ±1 % accuracy. Since about 1883 Otto Pettersson had refined the volumetric method by introducing means of eliminating volume errors caused by by heat of absorption. Therefore the former volumetric analyses show erratic values due to temperature dependence. The French school of analysing air had used sulphuric acid to dry the air. This had introduced another error of about 20–30 ppm too low, because of absorption of a fraction of the sampled CO_2 in the acid. The Belgian chemist W. Spring investigated this problem in detail in 1883. In the 20th century August Krogh, D. van Slyke, Y. Kauko and P. Schuftan introduced high precision volumetric, manometric, pH-metric and conductometric methods down to an error range of ±0.33 % in 1935 with an analysing time of some minutes. In 1938 the URAS (Ultrared Absorption Writer) gas analyser was invented by Friedrich Luft at BASF, the first continuous monitoring physical device later called NDIR, which is in use today, showing an error range of about ±0.1 %. (Beck 2007, 2008). Modern physical NDIR devices are calibrated against a manometric CO_2 determination.

Figure 3. Evolution of analytical methods for chemical CO2 gas analysis since 1800–1960.

The timeline of the methods in CO_2 gas analysis is summarized in Figure 3.

Kauko in 1934–37 did comparative analysis of well known analytical methods at that time (gravimetric, titrimetric, Pettenkofer, potentiometric, pH-metric, condensation) and Kauko et al. (1935) developed the most accurate CO_2 condensation method (±0.33 % valid down to CO_2-levels of 0.02 %) in 1935. According to Kauko all methods give comparable results within about ±3 %, the CO_2-levels concluded from the Pettenkofer method are somewhat lower than the exact levels

Table 1. Comparative chemical methods to quantify atmospheric CO_2-levels (Kauko 1934–1937).

pH method[1]	**potentiometric method**[2]	**condensation method**[2]	**Pettenkofer titrimetric**[3]	**gravimetric method**[4]
		0.017	0.0175	
0.036	0.031			
	0.065	0.065	0.069	
0.072	0.0721	0.0723		
0.087	0.088			
	0.122		0.1225	
0.144	1.42	0.143	1.43	1.422
0.18	0.18	0.189	0.172	
0.210			0.209	
0.239		0.236	0.241	
0.261		0.255	0.261	
	0.99	1.0	0.98	0.985
1.28		1.21	1.23	

Values in vol %; (1= Kauko 1934, 2= Kauko et al. 1935, 3= Hempel 1913, 4= Kauko et al. 1935).

Beside the methodical errors of the early gravimetric methods in weighting and drying the precipitate, which are difficult to reconstruct (de Saussure 1830), most of French measurement series, and those who use methods according to the Regnault-Reiset analysers show large errors because of CO_2 loss in drying ore setting free air by sulphuric acid (Reiset, Petermann, partly Montsouris) or a missing means of elimination heat of absorption in volumetric devices (Müntz plus other losses by using sulphuric acid). Other methodical errors derive from using too small volumes of air to analyse (early Pettenkofer methods, Frey, Heimann, Feldt).

All these errors have been quantified and corrected: Reiset: +20 ppm; Müntz: +20 ppm, Montsouris: +18.5 ppm 1877–june 1890, Feldt, Heimann, Frey: +40 ppm, Petermann: +20 ppm.

2.2 Analysis of vertical CO_2 profiles

The vertical CO_2-profiles are the key to estimate background levels from near-ground measurements. They are characterized by large seasonal fluctuation (SEAS) near ground on continents in non well-mixed environments and small variations in the higher troposphere or over sea surface (MBL) in well-mixed environments. Because all CO_2-sources are assumed to come from the lithosphere there is a physical connection from the ground to the higher layers. Figure 4 shows the most important globally CO_2-sources and sinks in the lithosphere-atmosphere boundary layer. Anthropogenic sources and others below 1 % of total emissions according to IPCC (Solomon et al. 2009) have been omitted. Within the atmosphere there exists a CO_2-gradient with a somewhat lower concentration and better mixing in the higher troposphere.

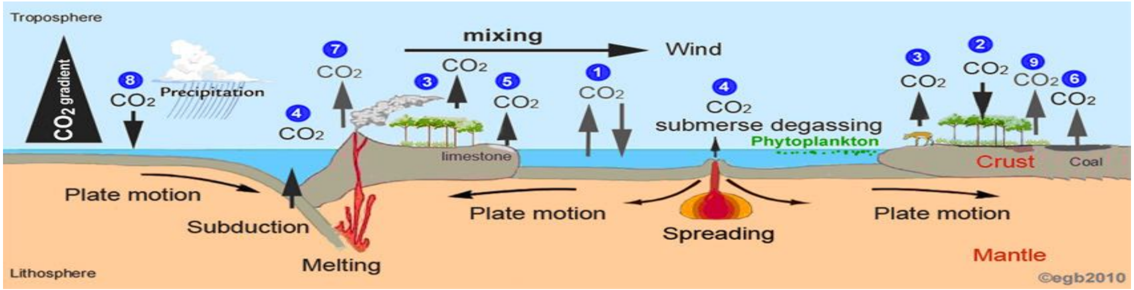
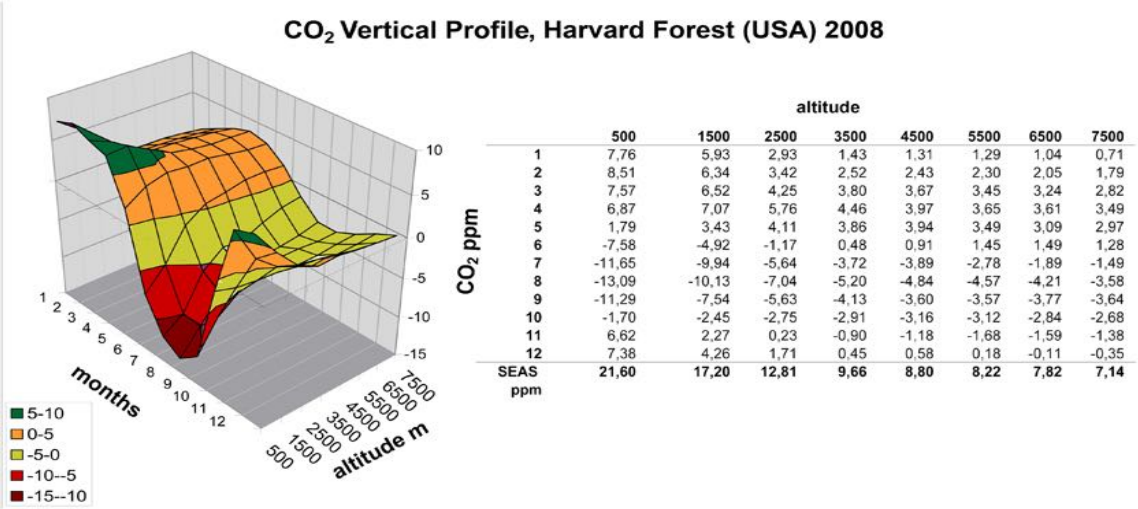


Figure 4. CO_2 sources and sinks in the boundary layer of the lithosphere-troposphere. 1: ocean degassing/ absorption, 2: photosynthesis, 3: respiration, 4: submerse geological degassing; 5: limestone weathering, 6: surface coal oxidation, 7: volcanic degassing and subduction degassing, 8: precipitation absorption, 9: soil respiration. CO_2 flux < 1 % of total emissions (IPCC) omitted.

The main globally effective controllers for CO_2-flux in the lithosphere/atmosphere system are the oceans (1) and the biomass (2, 3, 9). The phytoplankton in the surface layer of the oceans act as controlling agent for ocean bound CO_2.

The amount of geological surface flux of CO_2 from continents is greatly underestimated according to Mörner and Etiope (2002). Limestone weathering, surface coal oxidation and non-volcanic degassing are not quantified in detail in the IPCCs carbon cycle. Also the submerse fluxes in the oceans have not been quantified. (IPCC 2007). Local sources and sinks control local mixing ratios.



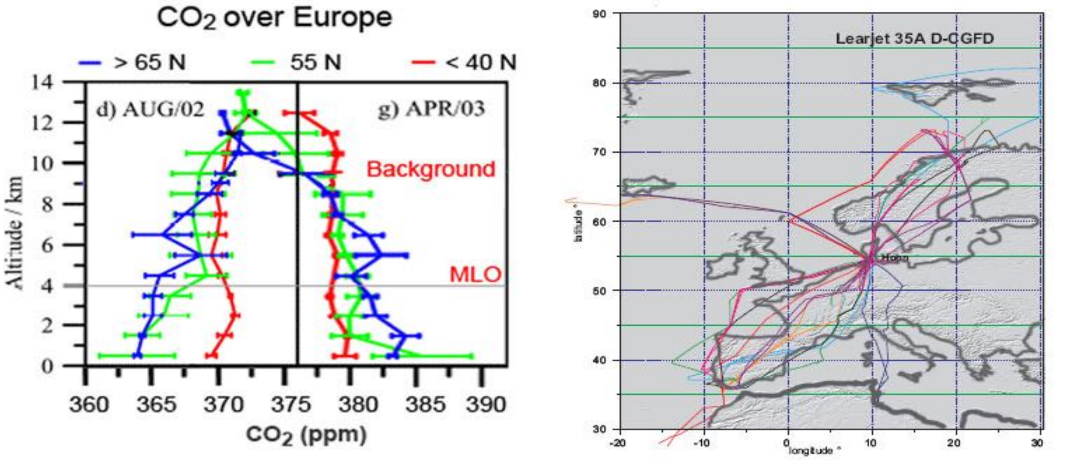
	500	1500	2500	3500	4500	5500	6500	7500
1	7,76	5,93	2,93	1,43	1,31	1,29	1,04	0,71
2	8,51	6,34	3,42	2,52	2,43	2,30	2,05	1,79
3	7,57	6,52	4,25	3,80	3,67	3,45	3,24	2,82
4	6,87	7,07	5,76	4,46	3,97	3,65	3,61	3,49
5	1,79	3,43	4,11	3,86	3,94	3,49	3,09	2,97
6	-7,58	-4,92	-1,17	0,48	0,91	1,45	1,49	1,28
7	-11,65	-9,94	-5,64	-3,72	-3,89	-2,78	-1,89	-1,49
8	-13,09	-10,13	-7,04	-5,20	-4,84	-4,57	-4,21	-3,58
9	-11,29	-7,54	-5,63	-4,13	-3,60	-3,57	-3,77	-3,64
10	-1,70	-2,45	-2,75	-2,91	-3,16	-3,12	-2,84	-2,68
11	6,62	2,27	0,23	-0,90	-1,18	-1,68	-1,59	-1,38
12	7,38	4,26	1,71	0,45	0,58	0,18	-0,11	-0,35
SEAS ppm	**21,60**	**17,20**	**12,81**	**9,66**	**8,80**	**8,22**	**7,82**	**7,14**

Figure 5. Vertical profile of CO_2 (deviations from 0) at Harvard Forest (USA), lat 42.54N, long -72.17E, measured by aeroplane at different altitudes of 500, 1500, 2500, 3500, 4500, 5500, 6500 and 7500 m.(data from NOAA Globalview-CO_2 2009)

From the NOAA Globalview-CO_2-sampling locations (NOAA 2009) I have chosen the vertical CO_2-gradient from the Harvard Forest site as an example for a typical continental location with vegetation at a typical latitude (lat 42.547N, long -72.17E).

Figure 5 shows the larger SEAS fluctuation near ground (500 m, 21.60 ppm) and the smaller variation of the background levels at higher altitudes (7500 m: 7.14 ppm). The SEAS average is nearly identical for 500 m: 0, 0.099 and 7500 m: -0.0055 (0.1 ppm difference).

CGER Monitoring of GHGs over Siberia supplies typical continental vertical data profiles (CGER 1996) and the work of Gurk et al. (2008) analysed vertical CO_2-profiles over Europe (land and



Source: Gurk et al., ACPD 8, 7315–7337, 2008 mod. ©egb2009

Flight path of the Learjet for all 8 SPURT measurement campaigns.

Figure 6. Vertical CO_2-profiles over Europe covering latitudes from 33 N to 79 N sampled by aeroplane during 2002–2003. (Gurk et al. 2008) showing the selected month with largest deviation. Horizontal bars are standard deviations for each km altitude.(flight path from Engel et al. 2006).

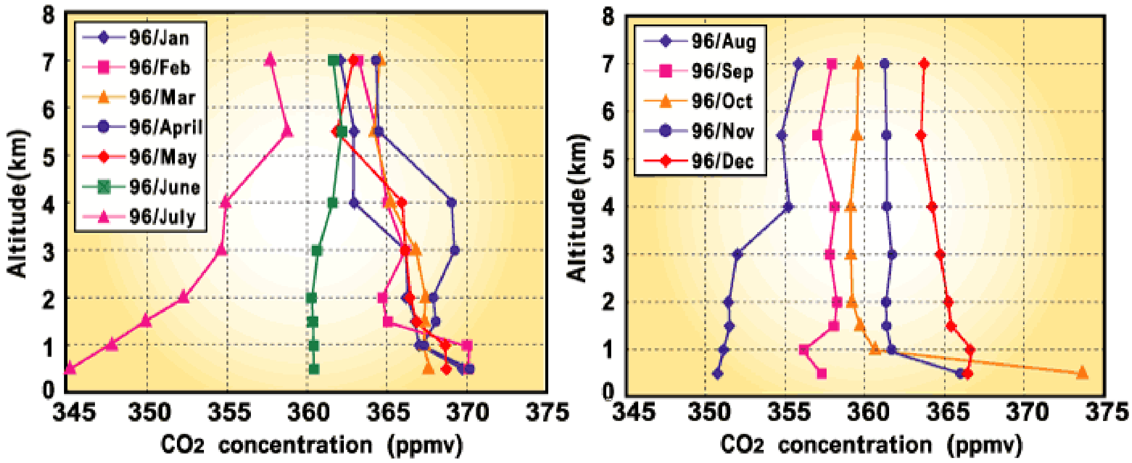


Figure 7. Vertical CO_2-profile sampled over Surgut, Siberia in 1996.

The seasonal variation in the boundary layer was max. 28 ppm. The variation at 4,000 m was 15 ppm. 14 ppm was the difference at the marine boundary layer calculated from Globalview-CO_2 (2008).

The annual average for CO_2 at 55 N at an altitude of 4 km was 373.5 ppm, the annual average at 500 m was 375 ppm, that means a difference of 1.5 ppm (-0.5 % deviation).

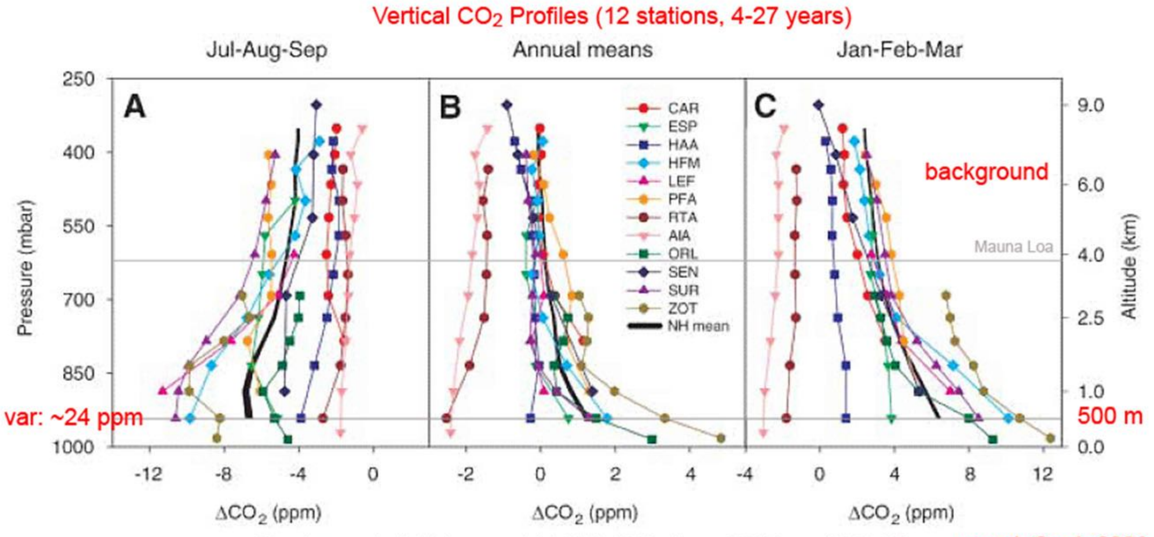
Over Surgut we found a seasonal range of 28.4 ppm at 500 m and 42.6 ppm extrapolated near ground. The Surgut data from 1996 show a summer deviation of –17 ppm for July compared to the global MBL background of 361.6 ppm.

Stephens et al. (2007) used average vertical profiles derived from aircraft during midday at 12 global stations at latitudes 40N to 40S with records extending over periods from 4–27 years averaged over different seasonal intervals. (See Figure 1.) The following locations have been chosen according to the NOAA Globalview-CO_2 Sampling locations

https://gml.noaa.gov/ccgg/globalview/

Table 2. Locations analysed by Stephens et al. (2007)

	Location	**Geographic position: Latitude, longitude**
1	Briggsdale, Colorado, United States (CAR)	lat. 40.37 N long. –104.30 E
2	Estevan Point, British Columbia, Canada (ESP)	lat. 49.58 N long. 126.37 E
3	Molokai Island Hawaii (USA) (HAA)	lat. 21.23 N long. 158.95 E
4	Harvard Forest, Massachusetts, United States (HFM)	lat. 42.54 N long. –72.17 E
5	Park Falls, Wisconsin, United States (LEF)	lat. 45.93 N long. –90.27 E
6	Poker Flat, Alaska, United States (PFA)	lat. 65.07 N long. –147.29 E
7	Orleans, France (ORL)	lat. 47.80 N long 2.50 E
8	Sendai/Fukuoka Japan (SEN)	lat. 33.39 N long. 130.21E
9	Surgut, Russia (SUR)	lat. 61N long 73 E
10	Zotina, Russia, Siberia (ZOT)	lat. 60.75 N long. 89.38 E
11	Rarotonga, Cook Islands (RTA)	lat. 21.25 S long. –159.83 E
12	Bass Strait/Cape Grimm Australia (AIA)	lat. 40.53 S long. 144.30 E



Stephens et al. Science , Vol 316, 22th June 2007, p. 1732-35

Figure 8. Vertical CO_2-profiles from 12 global stations derived from flask samples collected from aircraft during midday with records extending over periods from 4 to 27 years. (Stephens et al. 2007). Figure 1 of the paper was supplemented by the author. MLO = the Mauna Loa sampling station used as a reference. B has been calculated by subtracting the MLO data.

Figures 8 A and C show the seasonal variation (SEAS) at different altitudes and locations for July to September and January to March. Figure 8 B shows the annual averages of the deviation from the reference data sampled at Mauna Loa. The annual averaged difference of the seasonal variation to the Mauna Loa data in about 4 km altitude is about 2.3 ppm, near ground 8.6 ppm by graphical extrapolation. The average of the Northern Hemisphere data near ground shows a 2.56 ppm difference compared to the Mauna Loa reference at an altitude of about 4000 m. At 500 m we can observe a maximum seasonal variability of 27 ppm (from Globalview-CO_2) of all stations. Near ground by extrapolation this value extends to 30 ppm.

Table 3. Seasonal cycles near ground and background in modern data.

Location	**Max.Seasonal variation**	**ΔCO_2 background -500 m**	**ΔCO_2-background -near ground; extrapolated**
Europe 33N- 78 N, sea/land average	28 ppm		-
12 global stations 65N – 40S sea/land average	30 ppm	1.13 ppm	2.56 ppm
Surgut 60 N wetland	42.6 ppm	0.2 ppm	3.9 ppm

Analysis of vertical CO_2-profiles revealed that the yearly average of near ground values resembles the yearly average of the background level at altitudes higher than about 4 km at the same location within about 1 %.

2.3 Methods to estimate CO_2-background levels from continental near ground data: CWBA, CPBA, CSMBA, CDMBA

Local anthropogenic emissions, vegetation, soil respiration, geological soil degassing, wind and precipitations are the main influences on the CO_2-mixing ratios on continents near ground.

As the analysis of the vertical gradients at 500 m shows that annual averages are close to the mean, we can correct for the seasonal variations in the higher troposphere.

But near the surface (e.g. 1–50 m) mixing ratios can be much unsteady and CO_2-levels may vary in a wide range, dependent on local sources and sinks. Figures 9–12 show a range of up to 200 ppm at different locations inclusive some anthropogenic influence.

The Ameriflux network supplies a vast amount of high precision CO_2 and meteorological data from more than 130 sampling locations in continental environments with vegetation since 1996 at near ground levels (2–500m). About 30 data sources have been selected to work out methods for estimating annual background levels (Ameriflux 2009). Additionally the long time data series (1972–2008) from Schauinsland and data from some other stations have been used to test the CO_2-wind speed-background approximation (CWBA) developed by Massen et al. (2007), the CO_2- precipitation method (CPBA) (this paper), the summer minimum-background-approximation (CSMBA) (this paper) and the CDMBA, the daily minimum background approximation as proposed by C. Keeling (1958).

Background levels are characterized by well mixed conditions. Massen had shown that inland stations have a typical boomerang or sometimes a bi-lobe pattern investigating the CO_2-levels at rising wind speeds. At higher wind speeds (5–10 m/s) a quasi well mixed situation is achieved and the CO_2-levels represent the yearly average of the background level within a small error range (CWBA). By non-linear regression methods these CO_2-levels can be calculated. Equations from Massen (2004–2007 private communications) were applied to the data series in this paper:

$$CO_2 = A + B/X \tag{1}$$

X = wind speed or precipitation

Also other models are successful to fit the background levels:

$$Y = A * X^B \tag{2}$$
$$Y = A * e^{(B*X)} \tag{3}$$
$$Y = A + B*\ln(X) \tag{4}$$
$$Y = X / (A + B*X) \tag{5}$$

Since de Saussure 1810 (de Saussure 1830), it is known that precipitation lowers atmospheric CO_2-levels. This can be used to establish another tool to estimate CO_2 background levels per year within a small error range. Here we compare the CO_2-levels to the amount of precipitation. This emulates a quasi well mixed situation at high precipitation rates, because of washing out CO_2 from air to an equilibrium. The CPBA uses the physical effect of dissolution of CO_2 in water during times of precipitation. The remaining lower atmospheric CO_2 level is achieved faster than the CWBA during 2–3 mm precipitation. Again, non-linear regression models allow to estimate these lower CO_2-levels which are very close to the annual averaged MBLs.

Table 4. Data and error ranges for the CWBA and CPBA in Figure 9.

Stations	**MBL (NOAA) ppm**	**wind speed – MBL approx, ppm; w**	**precipitation – MBL approx, ppm; p**	**Error %**
Harvard Forest	MBL 2004= 376.76	377.67	378.78	w: -0.24, p: 0.53
Park Falls	MBL 2004= 376.76	382.57	383.66	w. -1.51, p: -1.79
Vaira Ranch	MBL 2004= 376.76	379.71	381.5	w. -0.77, p: -1.24
Hartheim	MBL 2003= 374.94	380.91		w: -1.57
Linden[1]	MBL 2008= 384.89	397.64		w: -3.2

Figure 9 shows typical examples for the validity of the CWBA and CPBA at continental locations with forest (Harvard Forest and Park Falls, USA), grassland (Vaira Ranch, USA), and rural area with agricultural use (Linden, Germany) and forest with anthropogenic influence (Hartheim, Germany).

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[1] Linden, HLUG, Hessisches Landesamt für Naturschutz, Umwelt und Geologie. https://www.hlnug.de/messwerte/datenportal

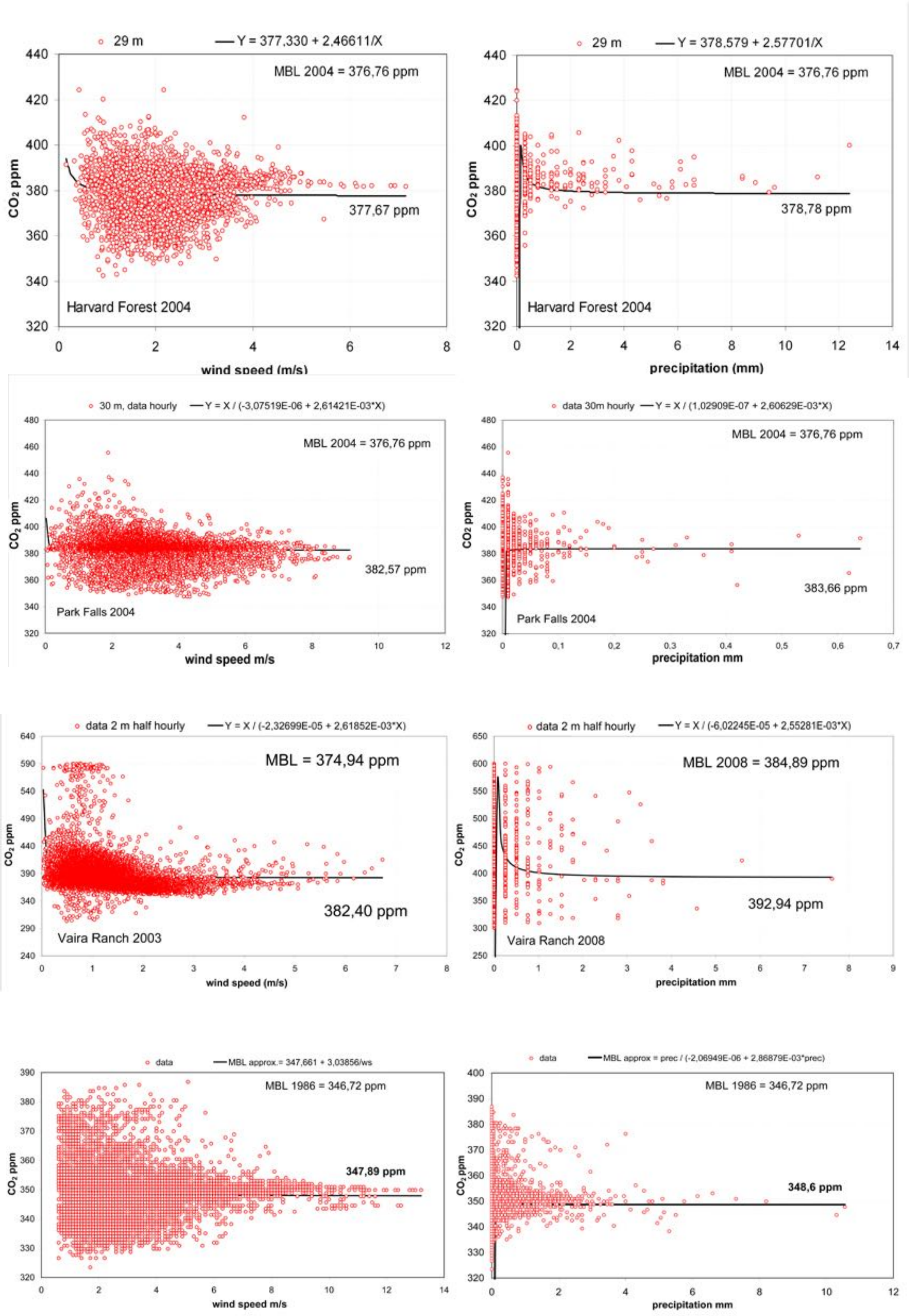

Figure 9. CO_2, wind speed (CWBA) and precipitation (CPBA) at Ameriflux station Harvard forest (lat 42.53N,-long 72.17E, 360 masl), Park Falls (lat 45.94N, long -90.27E, 480 masl), Vaira Ranch (USA, lat 38.4N, long -102.95E, 129 masl), Linden (Hessia, Germany: lat 50.32N, long 8.41E, 173 masl); Hartheim , (Rhine valley, lat 47.93N, long 7.62E, 206 masl), Black: linear regression and asymptotic fit from Massen models (1) and (5).

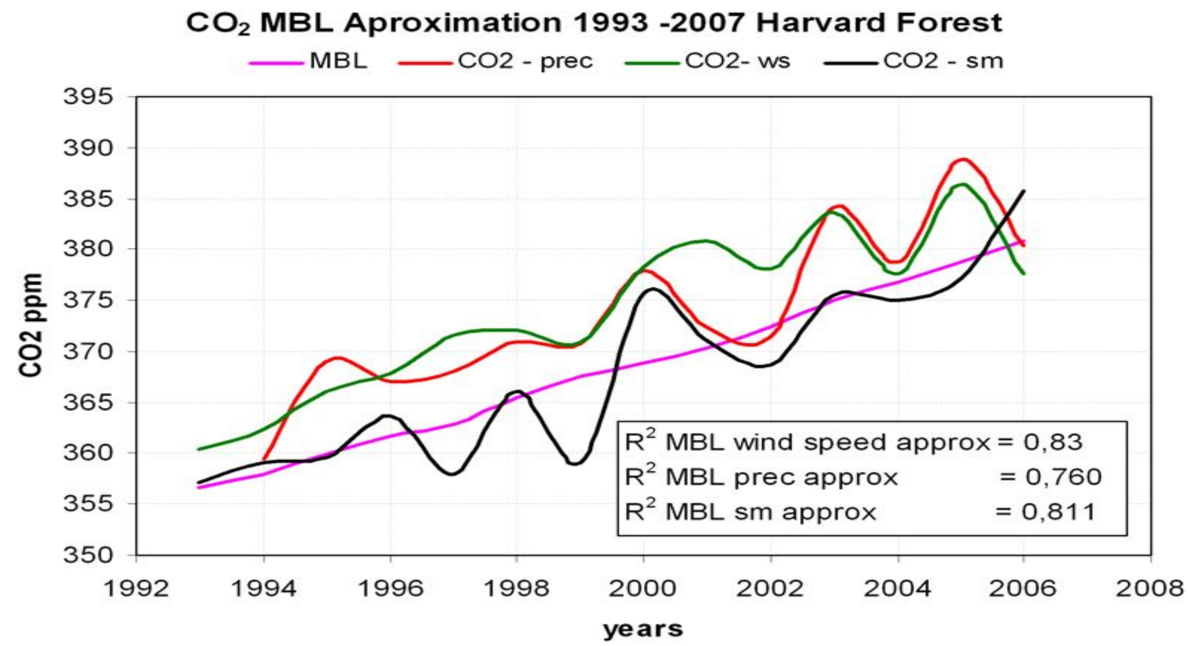

Figure 10. CWBA, CPBA and CSMBA at the Ameriflux station Harvard forest from 1993 to 2006 using hourly samples and their. coefficients of determination. (ws = wind speed, sm= summer minimum, prec=precipitation).

Typical forest locations as Harvard Forest and Park Falls show a very good approximation to the MBL annual averages. In contrast to CWBA and CPBA, the CSMBA does not reflect the result of a physical process to establish a quasi well mixed situation. It's based on the simple calculation of the seasonal average from the summer minimum average in June, July, August compared to the total average of the seasonal cycle which appears 2 times a year in April and October/November. Therefore the method is limited by the amount of anthropogenic bias of natural sources and sink meachanisms.

Figure 10 shows the application of CWBA, CPBA and CSMBA to the available data from the Ameriflux station Harvard forest from 1993 to 2006 using hourly samples. 2007 data have been omitted because of too few CO_2 data are available.

The coefficients of determination show the very good correlation of the methods. At Harvard Forest the CWBA fits in average 1.25 % and the CPBA 1.51 % to the MBL (1993–2006).

At grassland environments like Ameriflux Vaira Ranch (lat 38.4 long -120.9, masl 129 m) the CWBA and CPBA resulted in the same approximation limits as can be seen in forested stations. In 2004 the MBL of 376.76 can be approximated to +0.77 % by the CWBA and to +1.24 % by the CPBA. Please note the tri-lobe pattern as outlined by Massen and Beck (2011 in CWBA in their Figure 5 to the right).

The Hartheim and Linden station has been included because of strong local influences. The Hartheim data were collected in the very hot summer 2003/2004 at a Scots pine forest growing in the warm and dry southern upper Rhine plain. The location has mixed vegetation and anthropogenic influence. Schindler et al. (2006) indicated a netto CO_2 emission at that period. Nevertheless the CWBA results in an error of -1.57 % compared to the NOAA MBL.

The Linden station has strong agricultural and forestry influence. Using wind speeds from 1.5 m/s, regression results in 397.64 ppm, the asymptotic value is 396.45 ppm, which is an error of +3 %.

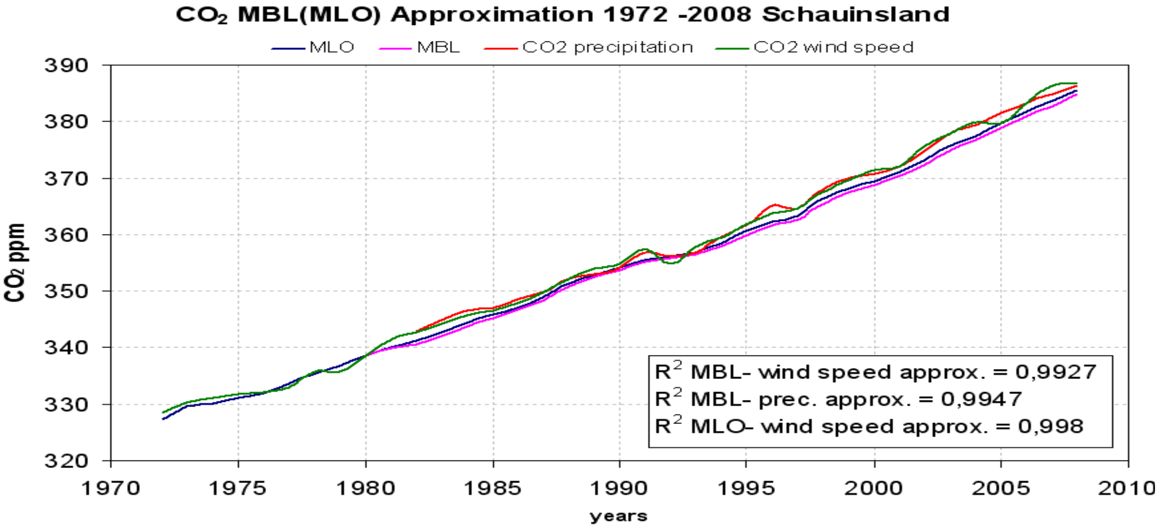

Figure 11. CWBA and CPBA applied to the CO_2 data series from Schauinsland, Germany, 1972–2008 lat (47.92°N, long 7.92°E, 1205 m masl), UBA (2009).

In Figure 11 the CWBA and the CPBA have been applied to estimate the global background levels from the 36 year long data series from Schauinsland (Germany) UBA (2009)[2]. The Schauinsland station (47.55N, 7.55E, 1205 masl) is surrounded by forests and meadows situated about 15 km South-Southwest of Freiburg and west to the Upper Rhine rift and the French boarder. Annual mean precipitation is about 1,780 mm. Annual mean temperature is about 5.6 °C. Prevailing wind is south westerly with anthropogenic influence mainly during nights. R2 are the coefficients of determination which show strong positive correlation and a very small error range within about 1 % despite of its vicinity to biogenic and anthropogenic sources and sinks. (Schmidt et al. 2003).

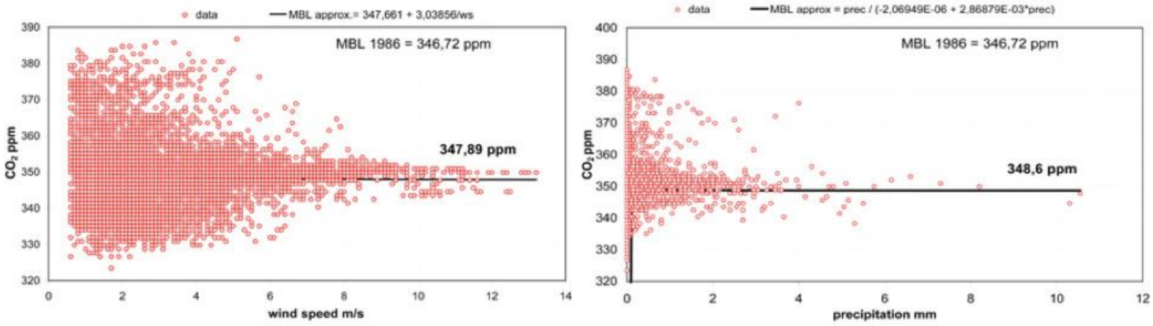

Figure 12. CO_2 wind speed (CWBA) and precipitation (CPBA) at the Schauinsland station during 1986 using ½ hourly samples. Black: asymptotic fit

Figure 12 shows a typical example of the CWBA and CPBA applied to the Schauinsland data 1986. Using one of the nonlinear regression models the CWBA results in 347.89 ppm, the CPBA in 348.6 ppm which is an error of +0.33 % and +0.54 % to the 1986 MBL of 346.72 ppm. The CPBA patterns for the period of 36 years at Schauinsland are nearly identical to the CWBA pattern and show that the MBL can be approximated within very small error range from about 4 mm precipitation.

C. Keeling (1958) had found in 1955 minimum daily CO_2 concentrations in the afternoon and observed the same at other locations. He suggested that this is a meteorological phenomenon. This can be used to check if this minimum value represents the MBL or background level in the higher troposphere.

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[2] Schauinsland data set supplied by Kerin Uhse UBA (Umwelt Bundesamt) 2009, outliers corrected.

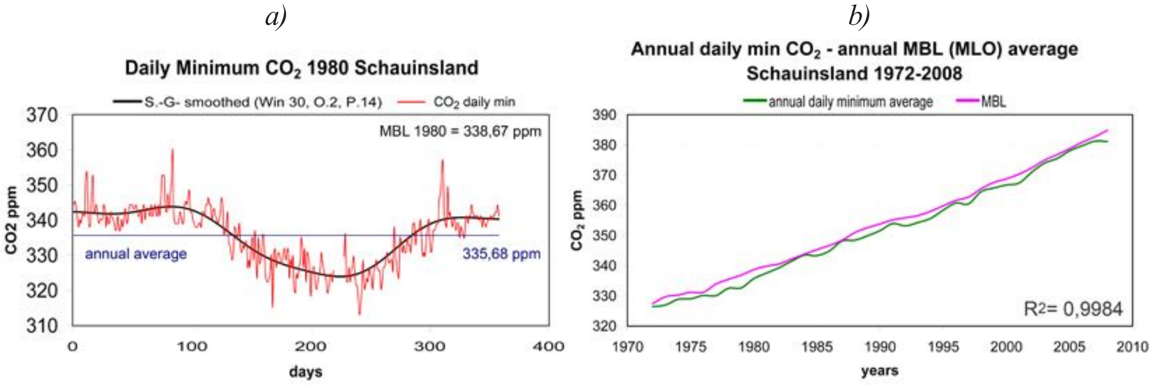


Figure 13a). Daily minimum CO_2-levels 1980 at Schauinsland (Germany) and b) annual average for daily minimum 1972–2008 compared to MBL/MLO annual average

Figure 13b shows the calculation of the daily minimum values for 36 years of annual data at Schauinsland (Germany). As an example the 1980 SEAS is presented as daily minimum smoothed by a Savitzky-Golay Filter (Win 30, order: 2, passes 14). This is shown in Figure 13a.

The daily minimum average (CDMBA = CO_2- daily minimum- background- approximation) at the Schauinsland station 1980 was 335.68 ppm which is only -0.88 % different from the MBL average of 338.67 ppm. The calculation of the CDMBA for the whole 36 years at Schauinsland reveals a very good correlation. Total average for the 36 years is 351.95 ppm and the MBL average 353.85, a -0.54 % difference. The smoothed seasonal amplitude is 17 ppm compared to the averaged 13.8 ppm deduced by Schmidt et al. (2003).

Additionally, the CWBA, CPBA and CSMBA were tested at dozens of other stations for example Mead Rainfall, Duke Forest, Howland Forest, UMBS, Niwot, AMT-Maine, ARM, WLEF, WKT, UCI, UMBS (USA, Canada), Wasserkuppe (Hessia), Hartheim (Germany) and more.

The MBL over typical forest and grassland environments can be modelled by CWBA and CPBA from near ground sampled data within 1 %. The CSMBA method approximates the MBL from near ground data within about ±1.5 %. At continental environments outside cities even with stronger anthropogenic influence the CWBA, CPBA and CSMBA are valid within about ±2.5 %. The CDMBA is also valid at mixed continental and marine environments showing errors around 1 %.

2.4 Estimation of atmospheric CO_2 background levels since 1800

We have applied vertical profile analysis, CWBA, CPBA, CSMBA and CDMBA to the historical CO_2 data series. The methods outlined in 2.3 has been applied to the historical data set compiled in the supplemental data files: CO_2MBL_1800–1960.xls and CO_2raw.zip. Daily data in CO_2raw.zip are the basis to calculate monthly averages in CO_2MBL_1800–1960.xls.

CO_2MBL_1800–1960.xls also contains the procedure to estimate the annual CO_2 background levels from these data for each selected station.

In contrast to the continuous sampling since 1958 the historical data have been collected at hundreds of different locations. Sampling periods differ in decades, years, months or days.

A lot of the historic measurement series have been collected only during seasons of the year mostly at summer time on continental stations. Therefore the presented 4 tools CWBA, CPBA, CSMBA and CDMBA have been applied when sampling period had fitted.

Using available meteorological and oceanographic data from each sampling location given in the historical literature to analyse air masses it is possible to estimate annual CO_2 background level within an error range of about ±3 % (chemical methods). A SEAS range is used between 12.5 and 18.5 ppm for years with MBL average around 315 and 380 ppm. At coastal stations winds from the sea are preferred for calculations, also CWBA, CPBA and CSMBA at continental stations or data sampled on rainy and stormy days.

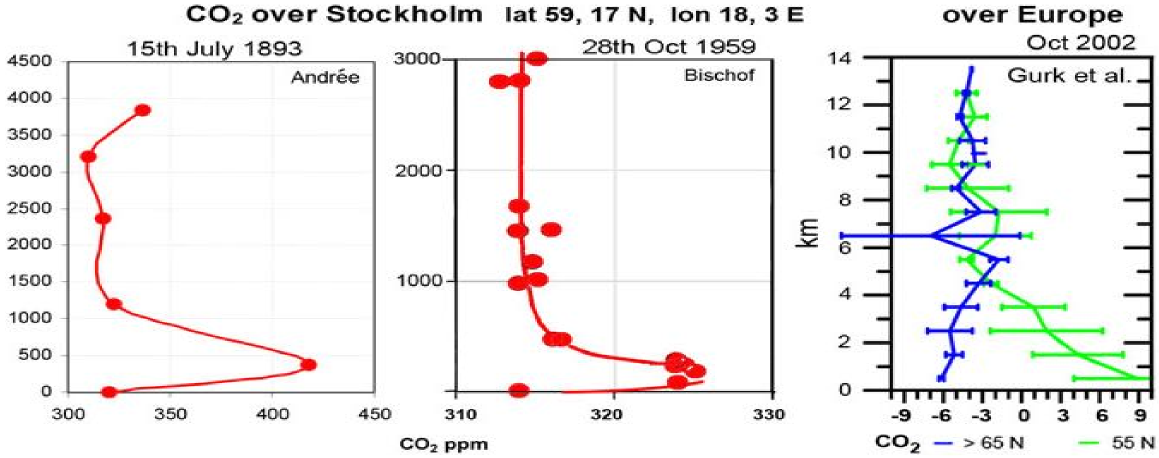


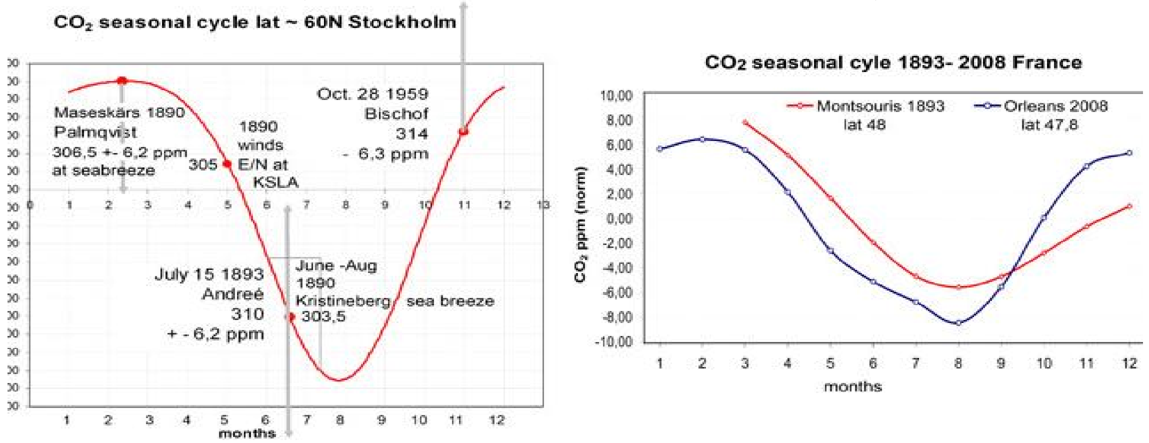
Figure 14. Vertical profiles of atmospheric CO_2 in the air over Stockholm 1893 (Andrée cited in Letts and Blake, Stockholm 1959 (Bischof 1960) and over Europe 2002/2003 (Gurk et al. 2008).
On the right: green lines= lat 55°N, blue= >65°N, modified from Gurk et al.

The following examples are typical for the estimation process:

2.4.1 Background estimation for 1890, 1893, 1935 and 1959

Available historical literature supplies 4 vertical profiles measured in 1893 by Andrée (Balloon near Stockholm, up to 3837 m; Petterson gas analyzer; error ~±1–2 %), 1911 by Wigand (Balloon Bitterfeld (Germany) to 9040 m; fractionated condensation method, according to Erdmann, error <±1 %), Kauko 1935 over Helsinki up to 1600 m (condensation method ±0.33 %) and Bischof 1959. near Stockholm as high as 3 km (NDIR <±1 ppm) (Letts and Blake 1899–1902, Stepanova 1952, Bischof 1960).

Figure 14 reveals the same CO_2-levels near ground as in the upper troposphere. The latitude of Stockholm is about 60°N so the assumed vertical profile is between the blue and green line which is near the yearly average. The amplitude of 16.5 ppm in Figure 12 used for the reconstruction of the 1893 CO_2 background level has to be reduced by about 10–15 ppm for the data at latitudes around 60 N.


a)

b)

Figure 15a). Calculated atmospheric background CO_2 in the air over Stockholm: 1893 Andrée cited in Letts and Blake (1899), 1959, Bischof (1960), and as a mix of MBL at Pallas and Baltic Sea. The contour (red line) is generated by mixing Pallas and Baltic Sea Globalview-CO_2 SEAS data and fitted according to the Loess Algorithm Cleveland (1993). b) Comparison of smoothed (Savitzky-Golay (Autosignal 2009)) measured SEAS at Montsouris 1893 and Orleans 2008 (Globalview-CO_2) (Montsouris data are without erratic January and February values).

According to the analysis in Figures 14 and 15, I estimate the CO_2 background level in 1893 measured over Stockholm to be 315 ±6.3 ppm. The MBL in 1959 must be 316.98 ppm (MLO+1). Bischof had measured 314 ppm in Oct. 28th, 1959. The October value must be 3.33 ppm above MBL. If we accept the MLO data, the Bischof value of 314 has an error of +6.3 ppm or +2 %.

A comparison of the SEAS measured at Montsouris in 1893 and the MBL SEAS from Globalview-CO_2 for Orleans in 2008, both at a latitude of around 48°N shows the good fit August as the summer minimum (Montsouris 307.3 ppm). The SEAS shows a good fit in 1893 and 2008 for despite of the difference in amplitude (about 13 ppm Montsouris) and levels (2008 about 380 ppm, 1893 about 310 ppm). The Montsouris data reveal a summer minimum of 307.5 ppm and a MBL level of 313 ppm assuming a 14 ppm amplitude.

Therefore, I conclude with a background summer minimum of 306.5 ppm in 1893 with a MBL average of 315 ±6.2 ppm for Stockholm 1893 measured by Andrée at latitude of about 60°N. The difference of about 2 ppm accounts for the latitude difference.

For the years 1889/90 we can use the marine and coastal data sampled at Kristineberg (Sweden), the island of Maseskärs (Sweden) and the continental data from Stockholm done by Augusta Palmqvist, Montsouris (Paris), Gembloux (Belgium) and Odessa (Ukraine). The most accurate temperature constant volumetric Pettersson gas analyser was used by Palmqvist (error 1–2 %). The Montsouris and Gembloux-data (Pettenkofer variant) have to be corrected by +18.5 and +20 ppm because of CO_2 absorption losses due to the management of water content and procedural similarities to the lossy Reiset gas analyser in drying acid before absorption in alkaline solution (Stanhill 1983; Beck 2007). Both data series show erratic SEAS all over the years with much too low values during winter time and lacking the typical continental amplitude of about 25–40 ppm. The Montsouris data show an about 20 ppm rise since July 1890 due to changing the procedure (Stanhill 1982).

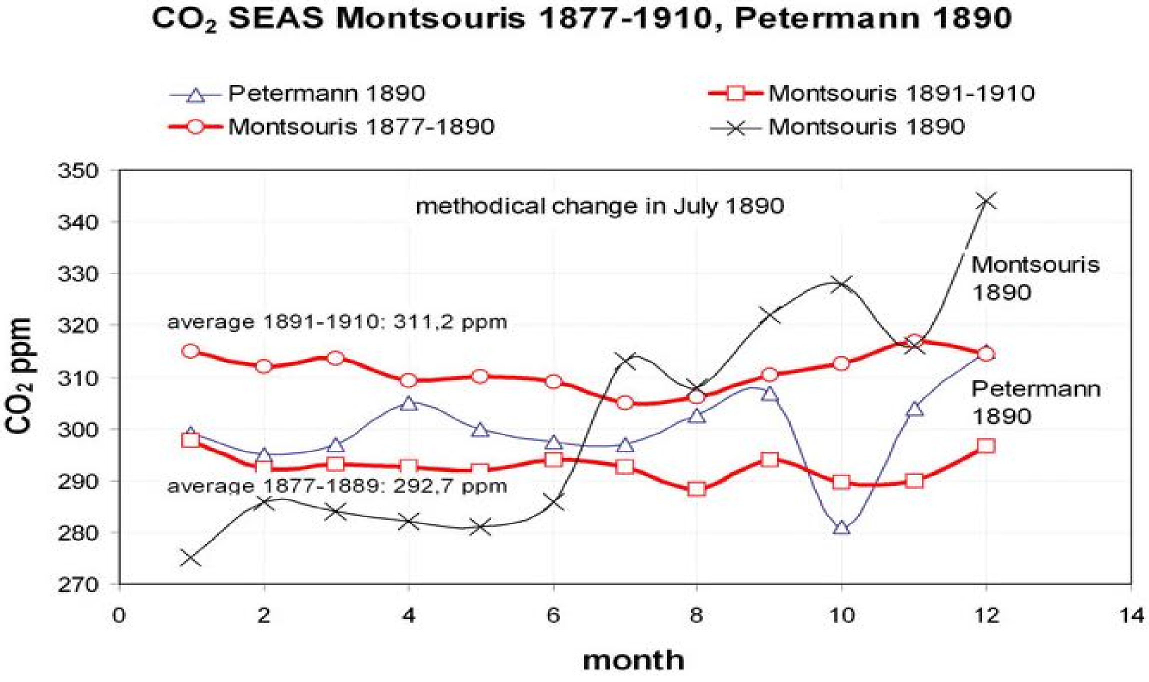


Figure 16. CO_2 SEAS analysed in Montsouris 1877–1910 and Gembloux (Belgium). Letts and Blake (1899). Methodical change in July 1890 leads to a shift up of 18.5 ppm in average SEAS (red). Erratic SEAS measured at Montsouris (black) and Gembloux (blue) 1890.

In 1890 Palmquist (1892) conducted 484 detailed air analyses at Stockholm (lat 59.2 N), Tromsø (Norway, lat 69 N), Maseskär (Sweden, Island, 7 km from the coast in Skagerrak, lat 58.5 N), Kristineberg (Skagerrak) and at the agricultural research station 1.5 km north of Stockholm KSLA) from 281 samples. CWBA and CPBA were not applicable to the Stockholm data (January to May average 323 ppm) because of the lack of precipitation observations in 1890 and only wind speeds of max. 2 m/s. Analysis of wind direction at the Stockholm station reveals lowest CO_2 in winds from eastern and northern direction with an average of 305.2 ppm from 21 samples in May. Winds from the sea at Maseskär island (Sweden) showed a CO_2 level of 306.5 in February which is taken as the reference for 1890 because of a marine location far from sources (stony island without vegetation), samples of air from the sea analysed with the most accurate volumetric temperature compensated equipment at that times (Petterson, 1–2 % accuracy), done by an experienced person.

Table 4 lists the data from 1890 at 7 different locations including 1125 samples. The Tromsø data show a weak influence of the mixed coastal/continental location as can be seen in the Odessa and Stockholm samples. The complete seasonal cycles measured at the continental stations at Paris and Gembloux had to be corrected because of the sulphuric acid loss error and other methodical errors. At Montsouris the method was changed in July 1890 resulting in a large shift upwards in the CO_2 level of in average 18.5 ppm which was added to the whole series after that date.

Callendar (1940, 1958) had listed the Montsouris and Gembloux data, also From and Keeling (1986) and Wigley (1983) had listed the Petermann data series, rejecting it because of the poor quality without discussing methodical aspects.

The amplitude of 16.5 ppm in Figure 15 used for the reconstruction of the 1890 CO_2 background level has to be reduced to about 10–15 ppm for the data at latitudes around 60 N.

Table 5. CO_2 in the atmosphere 1890 over Europe (1125 samples, 484 analyses in Scandinavia)

	CO_2 ppm	**samples**	**Station/ Author**	**lat**	**remarks**
1	306.5	38	Maseskär, Palmqvist	58.5	Sea breeze SW, W, NW; February
2	303.5	64	Kristineberg, Palmqvist	59.17	Sea breeze from June, July , August
3	314.4	23	Tromsø, Palmqvist	69	At stormy winds NW/SW
4	305.2	197	Stockholm KSLA, Palmqvist	59.2	Winds from E/N in May
5	304	-	Odessa (Ukraine), Lebedinzeff	46	Average March/April at rural area
6	302.1	365	Montsouris, Levy	46	SEAS average, corrected +18.5 ppm
7	300	365	Gembloux, Petermann	50.5	SEAS average, corrected +20 ppm

Estimation for the background level 1890: 302.5 ±6.2 ppm (Maseskär: 306.5 -6 ppm).

Figure 17 b) reveals that the average in about 1.5 km altitude over Europe in January 2002 for similar latitudes is close to 375 ppm. The annual average of CO_2 background at 4 km altitude from Figure 6 is 373.5 ppm which corresponds very well to the global MBL average of the years 2002/2003 of 373.7 ppm Globalview-CO_2 (2009). Figure 7 shows the same deviation of about 2 ppm from the average at similar latitude and altitude (e.g. Surgut). This indicates that in 1935 Kauko had measured the real background CO_2 in the air over Helsinki (lat 60.1 long 25E) of 361 ppm ±0.33 % in Dec. 7th and 375 ppm ±0.33 % at 1000 m in Feb. 20th over the clouds. For an estimation of the CO_2 background average for 1935 the modern seasonal averages listed in Globalview CO_2 at similar latitudes of Pallas Finland, lat 68N, Baltic Sea lat 55N, Zotino lat 60N and Shetland lat 60.1N from Globalview-CO_2 are helpful. Globalview MBL CO_2 data since 1980 are comparable to historic times because they exhibit about the same high atmospheric CO_2 range of about 360–380 ppm as the historic data to be evaluated.

The typical seasonal amplitude at the stations varies from 20.4 ppm at Zotino to 14.8 ppm at the Shetland station and 16.8 ppm at Pallas (lat 67.97 N, long 24.12 E).

The larger seasonal amplitude at Zotino (lat 60 N) is due to a typical continental location, the smaller Shetland cycle (60.1 N) corresponds to a typical marine station. The difference between the Baltic Sea seasonal cycle (lat 55 N) and Pallas cycle (lat 68 N) is <1 ppm in amplitude and summer minimum. Therefore, we conclude a seasonal cycle for the air over Helsinki as a coastal station of about 16.8 ppm.

For estimating the 1935 CO_2 background we need to check the available data:

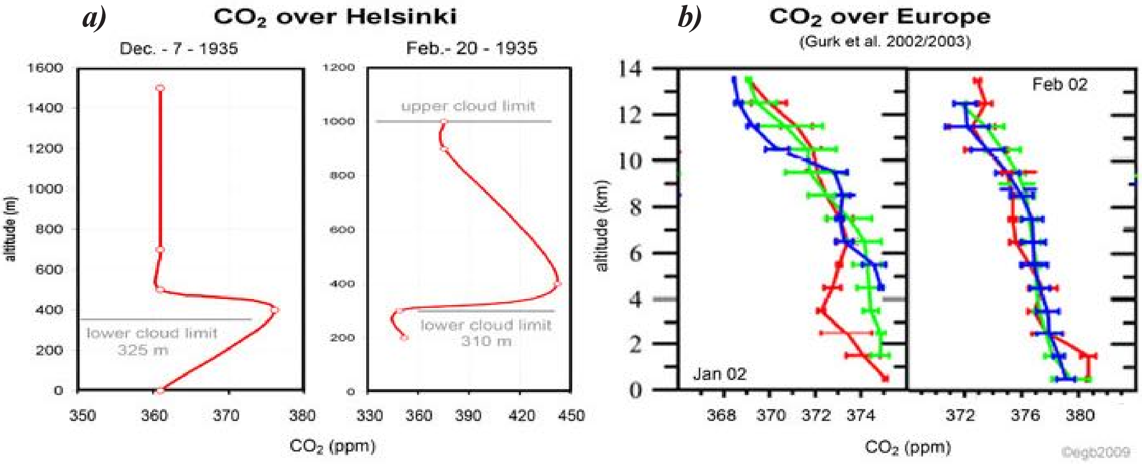


Figure 17a). Vertical profiles of atmospheric CO_2 in the air over Helsinki 1935 by Kauko and b) over Europe 2002/2003 (Gurk et al. 2008). On the right: green lines= lat 55N, blue= >65N, red<40N.

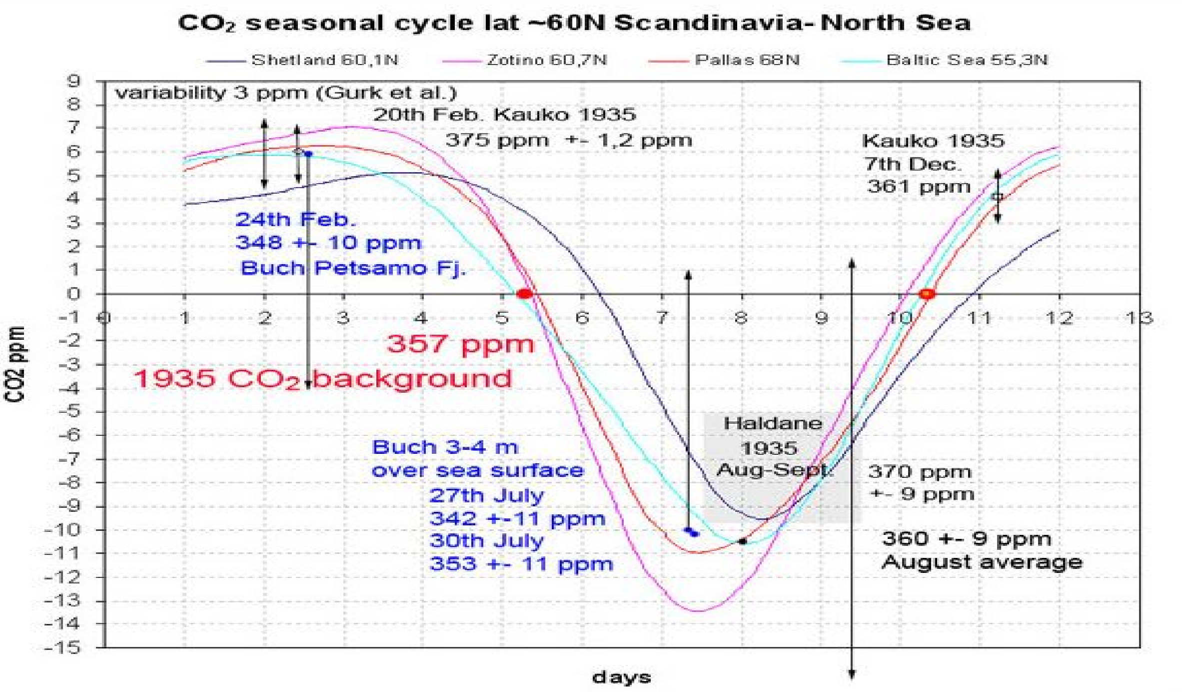


Figure 18. CO_2 seasonal cycles around lat 60N Scandinavia, North Sea (Globalview-CO_2 2008) and measured CO_2 around 1935 at various stations (troposphere, coast to Barent Sea, Scotland, Northern Atlantic Ocean. Arrows= error bars of sampling method. CO_2 data correct in time scale. Contours of graph fitted with the Loess algorithem.

Table 6. CO_2 in the atmosphere 1935/1936

1	361 ppm ±1.2 ppm	1600m altitude, Dec.1935, 1 km above cloud coverage, (Kauko); 3 x 361 ppm from 500 to 1600 m.
2	375 ppm±1.2 ppm	1 km altitude February, directly at the upper cloud limit (Kauko).
3	370 ppm ±9.2 ppm	near ground, winds from the sea Aug./Sept. average (Haldane).
4	360 ppm	average in August 1935 at Cloan (Scotland), rural station (Haldane).
5	348 ppm ±3 %	1935 at the Petsamo Fjord (Barents Sea) 24th February (Buch).
6	342 ppm ±3 %	1935 at Woods Hole USA coast and sea. (Buch).
7	353 ppm ±3 %	1935 Crossing the Atlantic Ocean (Buch)
8	>360 ppm	several CO_2 data 1934 (Buch over the Atlantic Ocean) and 1936 (Buch over the Atlantic Ocean Norway-Spitsbergen)
9	358 ppm	spring 1936 average, Duerst, Switzerland, rural location, only influence of vegetation)
10		Seasonal Cycles GlobalviewCO_2 lat N 55–68, modelled by the Loess algorithm.

During 1935/1936 another systematic analysis of air in Scotland is available by Haldane (1936). His volumetric gas analyzer was a standard in science for the first 50 years in the 20th century showing an accuracy of about 2.5 % (Beck 2007). The results of 1500 analyses, part of them using country air done at 1.2 m to 21 m above ground in Scotland (Cloan area, lat 56N and Ayrshire coast, lat 55N) since July 1935 are:

1. 153 daily analyses made at Cloan (Perthshire) during August 1935 were 320–400 ppm (average 360 ppm), 15 made at night showed 380–600 ppm. The whole range over ground was 210–440 ppm.

2. Samples of air blowing inland off the sea on to the coast of Ayrshire in August and September averaged 370 ppm of carbon dioxide and reflects the CO_2 rich air over the Firth of Clyde, North Channel and the islands there.

Buch did systematic analysis of air over sea in the northern and Arctic Atlantic Ocean from 1932 to 1936 Buch (1939a,b, 1948) during several travels by ship to Island, Spitsbergen, Woods Hole USA and New York. Furthermore, a data series sampled at Petsamo (Barents Sea) within 1.5 years is available. He used the gas analyser of Krogh and Rehberg (±3 % accuracy).

Duerst at mountains and shores of the Sea of Geneva (Switzerland) collected about 1,500 samples from 1936 to 1938. He used an optimized Pettenkofer method with ±3 % accuracy.

361 ppm reflects the December background, measured several times 1 km above clouds by Kauko. The February value of 375 ppm is the result of the influence of clouds. The 370 ppm measured by Haldane at the Scottish coast in August/September 1935 reflects the CO_2 enriched air of the coastal islands. Considering the error range and the fact that the difference continental/marine air at coasts are in the range of 10 ppm, the value has to be corrected by about 8 ppm. The average of 360 ppm at the rural Scottish environment reflects the continental summer minimum there. The value is near the correct CO_2 background within its ±2.5 error range of method and CSMBA(see below).

The Buch data supports the CO_2-levels around 360 ppm in 1935. His samples 3–4 m over sea show absorption by the ocean especially near the Arctic circle with lowest values. Samples at 30 m above sea surface had about 10 ppm higher values.

Inspection of adjacent years in 1934 and 1936 show similar data with higher values in 1936 . Buch had measured >368 ppm over the North Atlantic Ocean from Norway to Spitsbergen in 1936.

From the above analysis I conclude the average annual CO_2 background for 1935 is equal to 361 -4 ppm = 357 ±1.2 ppm and could have been measured according to the above evaluation of data at the end of May and October 1935 in the air over Helsinki.

2.4.2 Background estimation for 1920–1926

During 1920 to 1926 H. Lundegårdh analysed about 3000 air samples from June to September at Hallands Väderö, an island in Kattegat, southern Sweden using a gas analyser with optimized

Pettenkofer method to ±1 %. (Lundegårdh 1922).

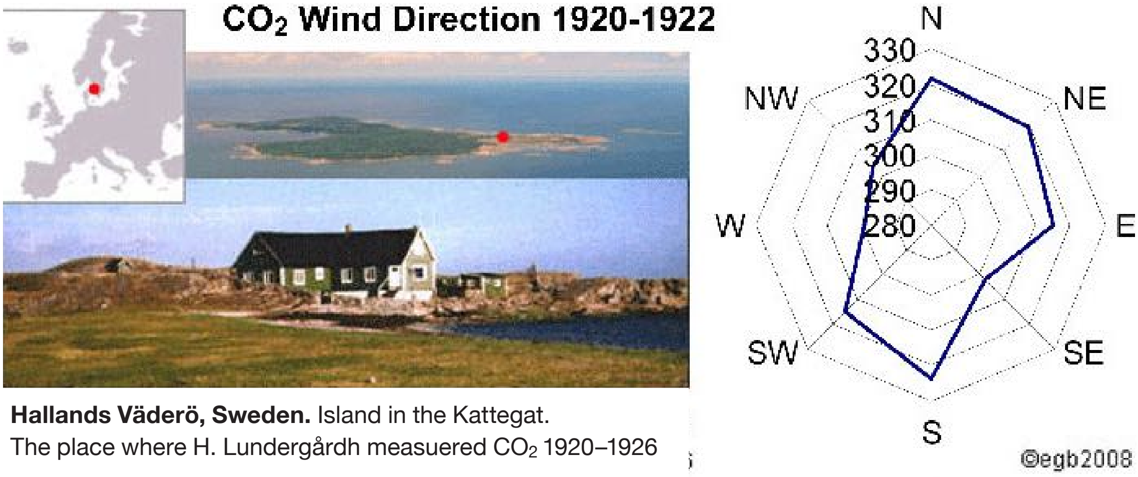


Figure 19. The ecological station of H. Lundegårdh 1920–1926 analysing air samples in the summer at Hallands Väderø (southern Sweden). The CO_2 wind direction 1920–1922 is presented at the right. S = south direction (photo ©http://www.shvn.se/); 8 masl; lat: 56° 25.8'N long 12° 34.3'E. (Lundegårdh 1924, 1949).

Wind analysis in Figure 19 shows that winds from the open sea (W, NW) have the lowest CO_2 concentration in summer (about 305 ppm). Air masses from the Northern and North-Eastern forests on the island provide higher CO_2 mixing ratios (about 320 ppm). Data in 1923 show mainly winds from the western direction. The station can be categorized as marine and coastal.

Table 7. CO_2 data (Hallands Väderö summers 1920–26).

	1920	**1921**	**1922**	**1923**	**1924**	**1925**	**1926**
Temperature °C	19.1	17.3	16.5	15.4	16.4		
averaged CO_2 ppm, measured	329.5	303.1	284.3	300.0	331	312.5	320.0
reconstructed annual CO_2 background	316	306.8	290.1	301.8	319.9	318.3	321.9

From Lundegårdh (1924, 1949); recalculated data.Data June –August 1920–1926
**data recalculated by using : ppmv= (mg/m3)(273.15+C°)/ 12.187(44.01)*

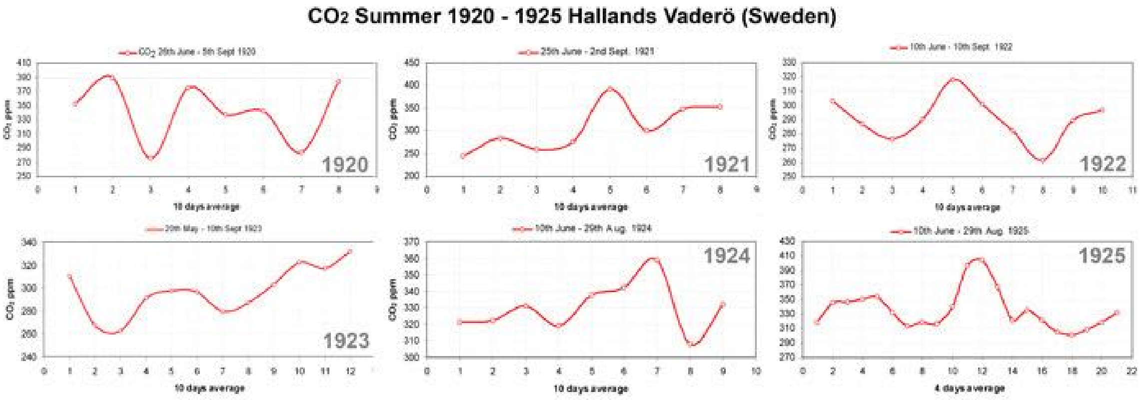


Figure 20. CO_2 in air at the shore of Hallands Väderø during 1920 to 1925 (10 days average June 29–Sept 2).

The June/July data usually show lower CO_2 than the second half of the summer. CPBA for summer 1921 estimates 306.76 ppm as the MBL for 1921 at Hallands Väderö.

The CO_2-levels through these years are characterized by a drop of > 20 ppm in the summer 1923 and reconstituting the former levels within 3 years. At the same time SST and temperature also drop down in the North Atlantic Ocean at lat 45–80N and long 0–15E at about 1.5 °C in average (see Figure 21 left). Figure 21 shows the evolution of three parameters in time as summer averages (June-July-August). SST is taken from COADS (KNMI), air temperature from Copenhagen (NCDC 2009) and CO_2 at Hallands Väderø according to Lundegårdh (1924, 1949).

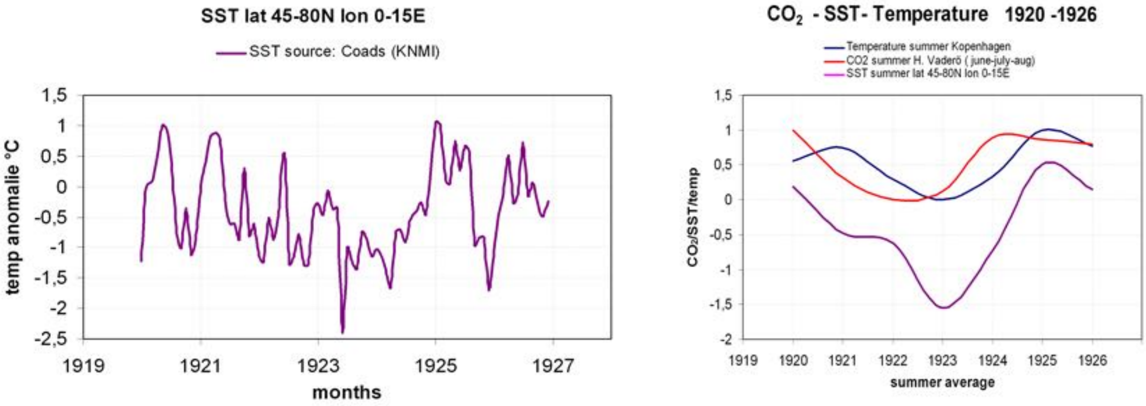


Figure 21. Monthly SST (left violet; COADS, KNMI), right: Copenhagen; summer temperature (blue: NCDC 2009), CO_2 (red): at Hallands Väderø, Kattegat (Lundegårdh (1924, 1949), SST summer (violet). All curves are anomalies.

Cross correlation calculation reveals a lag of 1 month of CO_2 behind SST. In Figure 22 one can see that the CO_2-levels mimic the SST contour with a time lag of about 1 month.

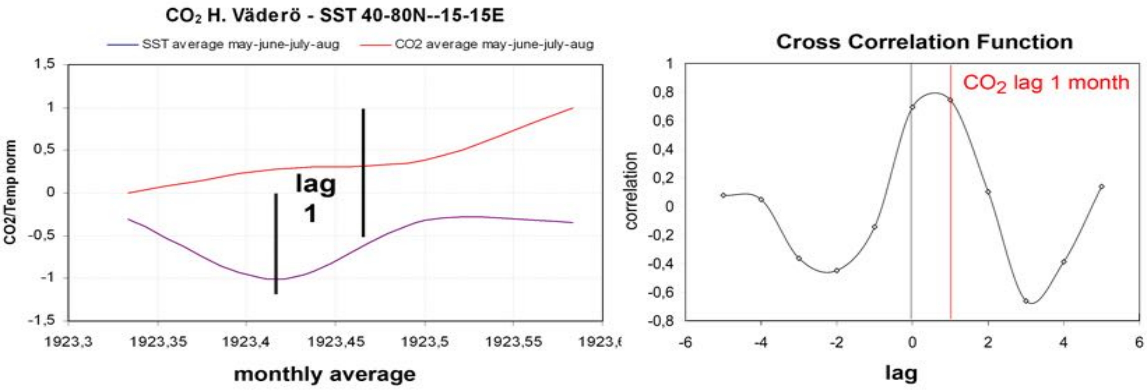


Figure 22. Time lag of CO_2 with respect to SST in 1923 at Hallands Väderø as monthly averages (left); cross correlation calculation (right).

In order to reconstruct the annual CO_2 background mixing ratios from 1920 to 1926 it is necessary to use the seasonal cycle data analysed at a nearby modern station, Baltic Sea (55.35°N, 17.22°E, 28 m; Globalview-CO_2, 2008).

In the summer of 1921, B. Schulz collected sea- and 25 atmospheric CO_2-samples during a cruise by the explorating ships Poseidon in the North Sea, and Skagerak (16th–31th July), and (24th August–17th September) in the Baltic Sea (Schulz 1922). He reported that the surface layers of the sea were always super saturated by CO_2 especially in the Baltic Sea. The Poseidon cruised in the Kattegat along the coast of southern Sweden (Hallands Väderø) and the Baltic Sea. The samples were analysed in the laboratory of the Deutsche Seewarte using the Krogh gas analyser (accuracy ~±2 %).

Table 8. CO_2 level in samples during the cruise of the Skagerak, Kattegat, and Baltic Sea July–Sept 1922.

Samples	1	2	3	4	5	6	7	8	9
CO_2 ppm	305	290	320	300	275	290	310	290	290

Average: 297 ppm; average sea surface CO_2 343 ppm (Schulz 1922).

From these data and the average in 1921 at Hallands Väderö we calculate a summer minimum of 301 ppm using the average of 5.1 (+1.7 % ppm from Table 3 and 6.5 ppm as the summer minimum from the seasonal cycle (Baltic Sea) as an addition, resulting in 306.8 ±4.6 ppm (error level from method 1.5 % method) as the background level in 1921 over Hallands Väderö. The low CO_2-levels in 1922 are estimated as follows:

CWBA summer 1922: 283.9 ppm; CPBA summer 1922: 282 ppm. Lundegard average 284.3 +5.8 ppm = 290.1 ±2.9 ppm.

The levels in 1920 show very high outliers around the 10th, 26th July and 3rd August. Eliminating these values and considering the very different SST values during this year the estimation for 1920 at Hallands Väderø = 318.3 ppm summer minimum +5.1 = 323.4 ppm ±3.2 ppm (error level from method 1 %).

In 1923 the winds came mostly from W/NW at Hallands Väderö with CO_2-levels just below 300 ppm. The summer minimum from data is 296 ppm, CPBA for the summer results in 298.9 ppm, CWBA is 283.85 ppm, so we estimate the CO_2 background level to 296 +5.8 ppm = 301.8 ±3 ppm.

The air in 1924 at Hallands Väderö show higher CO_2-levels again accompanied by a rise of the SST by about +2 °C in average in the Baltic Sea and Kattegat. Correcting outliers around the 22th, 48th, 61th and 66th day leads to 324.9 +5.8 ppm = 330.7 ±3.3 ppm. Summer CPBA is 337.5 ppm. The June average by Lundegårdh was 319.7 ppm, Meinecke at Eberswalde had measured 320 ppm after rain. So we had to correct the CO_2 background for 1924 to 319.9 ppm.

In 1925 E. Rheinau analysed air near ground and around Davos, Switzerland (1600–2500 m elevation) using the Petterson/Sonden gas analyser at the meteorological station Davos in August 1925, Rheinau (1926). 154 samples have an average of 293 ppm. A lot of days show fog and rain, at clear days levels are well beyond 310 ppm. The data are ignored because of too large uncertainties.

Lundegårdh (1949) lists 312.5 ppm in his table 45 p. 32. Digitalization from his graph p. 33 results in an average of 335.5 ppm, outlier and temperature corrected to 329.5 ppm. Because the SSTs are still down in 1925 we use 312.5 +5.8 = 318.3 ppm.

Table 9. 1926 summer observations.

Station, author	**CO_2 ppm**
Lundegårdh, Hallands Väderö	320
Krogh et al. (1929), Copenhagen, Greenland	318.5, 312, 310, 328
Wattenberg (1933) over sea surface in Atlantic Ocean	313.4
Average	**316.9**

CO_2 background estimation for 1926: 316.9 +5 ppm summer minimum = 321.9 ppm as 1926 CO_2 background level.

2.4.3 Background estimation for 1939–40

Figure 23 presents a re-read of the data of W. Kreutz compared to the data presented in Massen and Beck (2011). The CWBA difference is (392.6–390) = 2.6 ppm = 0.7 %.

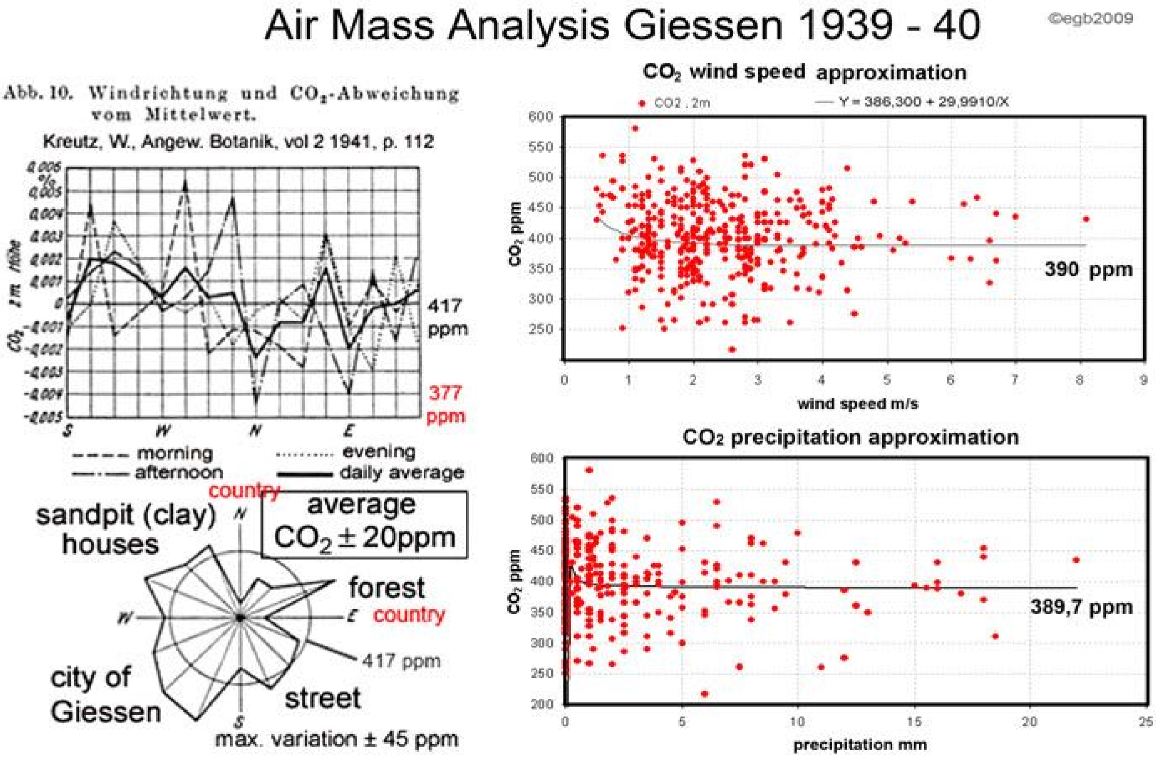


Figure 23. Air mass analysis using Giessen data from 1939–40. CO_2 variation sampled every 90 minutes 2 m above ground. On the left the air mass analysis done by W. Kreutz 1940, on the right the CWBA and CPBA at Giessen sampling station. The Massen model and asymptotic fit shown as grey line.Data from Massen and Beck (2011).

The approximated background level at Giessen 1939/40 by CWBA was 390 ppm and by CPBA it was 389.7 ppm, which is in good agreement to the air masses with the lowest CO_2-levels blowing from the north and east (rural area) at that times as deduced by W. Kreutz in 1942 of about 377 ppm. (Beck 2007). The standard deviation is 6.47. The CSMBA (June-July-August) cannot be calculated because the data series starts in August 1939. The August average is 338.5 ppm and the average for August/September is 347.8 ppm. T´he October average was 386 ppm.

The accuracy of the volumetric gas analyzer used (RICO C) was ±1.5 % (Schuftan 1933, cited in Beck 2007). So we can calculate ±1.7 % (CPBA) from the average (389.7+390)/8 + 386/4 +377/2= 382.4 ±2.5 %. This CO_2 level must be therefore considered as CO_2 background level at Giessen in 1939/40.

In a recent paper by Massen and Beck (2011) we presented a CWBA for additional historical data series e.g. at Vienna 1957/58 by F. Steinhauser at the meteorological station Hohe Warte estimating a MBL of 324 ppm.

3. Results and Discussion

The analysis presented above results in the estimation of eleven MBL approximations as examples for the whole data set of historical near ground sampled CO_2 values.

Table 10. Some MBL examples.

Year	Location	MBL estimate		Year	Location	MBL estimate
1890	Scandinavia ~60 N	302.5 ±6.2 ppm		1924	Hallands V. 55 N	319.9 ±3.3 ppm
1893	Stockholm 60 N	315 ±6.2 ppm		1925	Hallands V. 55 N	318.3 ±3.2 ppm
1920	Hallands V. 55 N	316 ±3.2 ppm		1926	Hallands V. 55 N	321.9 ±3.2 ppm
1921	Hallands V. 55 N	306.8 ±3 ppm		1935	Helsinki 60N	357 ±1.2 ppm
1922	Hallands V. 55 N	290.1 ±2.9 ppm		1939	Giessen 48N	382.4 ±9.5 ppm.
1923	Hallands V. 55 N	301.8 ±3 ppm				

Applying the methods outlined above to all available historical data listed from chemical CO_2-analysis listed in table 1, 134 yearly averages have been calculated representing the estimated CO_2 background levels. Since 1870 to 1960 errors had been estimated to ±0.33 -3 % or max. ±9 ppm referred to a CO_2 level of 300 ppm accepted as average through the two centuries.

Table 11 lists the annual CO_2 MBL estimations since 1826 to 1960 from data compiled in the files *CO2_MBL_1800-1960.xls* and *CO2_raw_1800-1960.pdf.*

Table 11. Annual CO_2 MBL estimations from 1826 to 1960 from direct measurements

1	2	3	1	2	3	1	2	3	1	2	3	1	2	3
1826	364.50	72.90	1856	335.20	33.52	1886	299.00	8.97	1916	323.40	9.33	1946	336.00	47.04
1827	359.00	71.80	1857	337.00	33.70	1887	302.80	9.08	1917	321.60	10.20	1947	338.00	47.32
1828	358.00	70.80	1858	335.20	33.35	1888	300.00	9.00	1918	317.00	10.46	1948	328.40	45.98
1829	339.00	67.80	1859	333.50	33.50	1889	300.80	6.02	1919	320.20	10.44	1949	316.00	9.48
1830	339.90	67.98	1860	331.80	33.30	1890	302.40	6.05	1920	323.40	10.43	1950	308.85	9.27
1831	340.80	102.24	1861	330.10	33.10	1891	308.10	9.24	1921	305.80	3.07	1951	314.50	9.44
1832	341.70	102.51	1862	328.40	32.90	1892	311.30	9.34	1922	289.40	2.90	1952	314.50	6.29
1833	342.60	102.78	1863	326.70	32.70	1893	310.00	6.30	1923	305.10	3.02	1953	318.90	6.36
1834	343.50	103.05	1864	325.00	32.50	1894	317.00	9.51	1924	330.00	3.20	1954	321.90	6.44
1835	344.40	103.32	1865	310.00	31.00	1895	305.00	9.15	1925	321.30	3.18	1955	318.80	6.38
1836	345.30	103.59	1866	301.20	9.04	1896	313.80	9.41	1926	321.90	3.22	1956	320.50	6.41
1837	346.20	103.86	1867	300.00	9.00	1897	312.00	9.36	1927	327.00	5.89	1957	320.20	6.43
1838	347.10	104.13	1868	298.00	8.94	1898	317.30	9.52	1928	327.00	6.54	1958	318.40	6.37
1839	348.00	174.00	1869	290.00	8.70	1899	313.60	9.41	1929	328.00	6.56	1959	317.80	6.36
1840	347.00	173.50	1870	291.00	8.73	1900	307.50	9.23	1930	328.00	6.56			
1841	341.50	170.75	1871	300.00	9.00	1901	311.50	9.35	1931	329.25	6.58			
1842	336.00	168.00	1872	313.90	9.42	1902	305.90	9.18	1932	326.40	4.90			
1843	325.00	162.50	1873	311.00	9.33	1903	306.50	9.19	1933	328.00	4.89			
1844	328.50	164.25	1874	317.50	9.52	1904	305.50	9.17	1934	331.00	4.96			
1845	332.10	166.05	1875	324.00	9.72	1905	307.00	9.21	1935	357.00	5.36			
1846	335.60	167.80	1876	311.00	9.33	1906	306.00	9.18	1936	349.50	6.99			
1847	339.30	169.65	1877	298.00	8.94	1907	307.10	9.23	1937	354.00	7.08			
1848	319.90	63.98	1878	303.50	9.11	1908	306.50	9.26	1938	354.00	7.08			
1849	330.00	66.00	1879	309.90	9.30	1909	303.30	7.75	1939	382.40	5.74			
1850	330.00	66.00	1880	300.00	9.00	1910	307.80	7.72	1940	376.50	5.65			
1851	323.60	64.72	1881	309.00	9.27	1911	308.20	7.69	1941	376.00	5.64			
1852	325.30	65.06	1882	312.00	9.36	1912	318.50	15.21	1942	379.50	15.18			
1853	328.30	65.66	1883	312.00	9.32	1913	329.00	9.24	1943	383.00	15.32			
1854	331.60	66.32	1884	312.00	9.36	1914	327.15	9.35	1944	367.30	14.69			
1855	333.50	66.70	1885	303.50	9.11	1915	325.30	9.46	1945	352.00	14.08			

1: year; 2: CO_2 MBL estimation; 3: ±error (ppm) of method

Figure 1 shows about 75 % of the data as monthly averages within a latitude range of 40–80N modelled by a distance weighted least squares fit algorithm. A clearly enhanced CO_2 concentration around 1940 over the whole latitude range can be seen with CO_2 maximum in 1942.

A continuous time series using estimations from Table 11 from 1826 to 1960 is presented in Figure 24.

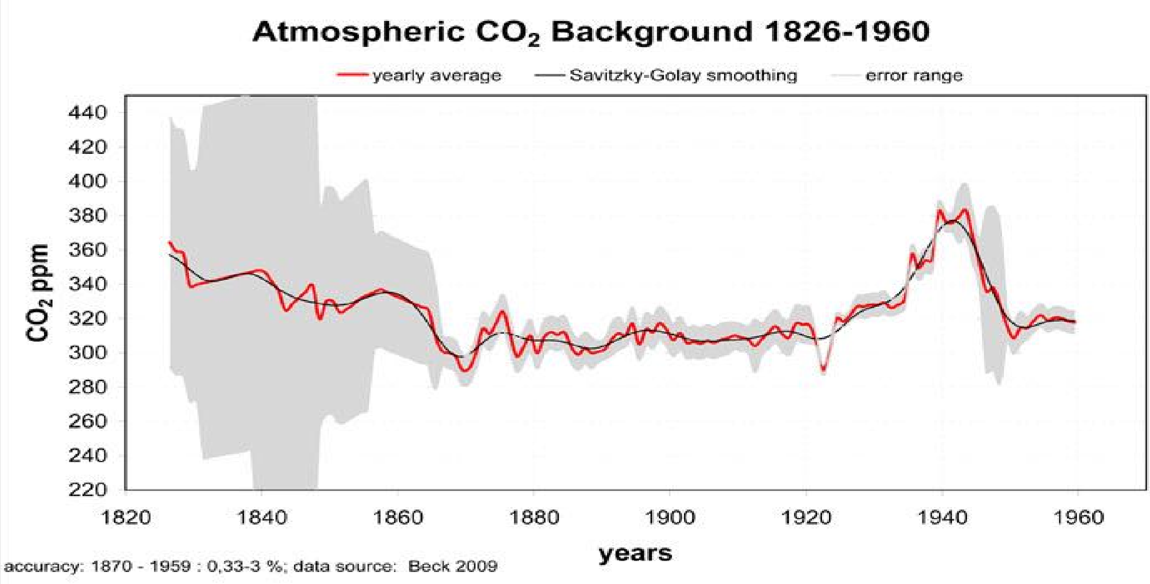


Figure 24. Atmospheric CO_2 background level 1826–1960. CO_2 estimates from directly measured data (red); black line smoothed by a Savitzky-Golay filter, grey area = estimated error range. Savitzky-Golay smoothing parameters (Autosignal): width of the moving window: 5, order: 2; passes: 3. Data from table 11.

Because of some gaps in the annual time series and uncertain data, 40 interpolations are necessary since 1826 (14 since 1870). 44 % coastal data or data from the sea or the higher troposphere since 1870 have been used. The CO_2 maximum in 1942 is based on the work of >25 authors and locations and >60 000 samples within about 20 years from 1930 to 1950. Additionally, the direct measurement of the background level of 357 ppm in 1935 over Helsinki by aeroplane, Kauko (1935) using the most accurate gas analyser in history of chemical methods (0.33 %), supports the existence of 1942 CO_2 maximum. It is most remarkable that literature reveals CO_2 enriched air coming from the sea at several stations when sampling at the coast (Haldane North Sea, Buch 1932–1936 Barents Sea, Northern Atlantic) or over warmer ocean currents in the Northern Atlantic (Buch1932–1936). This suggests the Northern Atlantic Ocean as the source of the enhanced CO_2-levels.

Using the Savitzky-Golay smoothed data (see Figure 24), the contour of the reconstructed MBL of CO_2 from directly measured data show maxima around 1858 (335.2 ppm) and 1941 (377.1 ppm) in contrast to the monotone rising CO_2-levels reconstructed from ice cores (IPCC). Several small oscillations of the curve prior to the continuously sampled CO_2 since 1958 are within error range of ±3 % since 1870:

Using a Savitzky-Golay smoothed curve with a wider moving window width of 7 to correct problems around 1880 and 1930 we can observe a periodic cycling with maxima at 1836: 343.9 ppm; 1857: 334.2 ppm; 1878: 309.7 ppm; 1896: 312 ppm; 1915: 320.7 ppm; and 1941: 371.4 ppm.

The average period is 21 years (21, 21, 18, 19, 26 years). The sharp drop in 1922 and the changing period length at those times points to phase change in the time series which supports the findings of Yndestad in the NAW (Northern Atlantic water temperature) (Yndestad et al. 2004). He found strong evidence for the lunar nodal cycle and its harmonics as the controlling forces behind the northern and Arctic climate. At around 1920 there is a phase-reversal of the 18-year temperature cycle in the NAW. Yndestad also identified 55 years as a dominant wavelet cycle and third harmonic of the 18.6 yr lunar nodal cycle. By calculating the best low-frequency harmonic fit using Lyubushin (2009) software of the data since 1870 we get a period of 55.17 years (see Figure 25).

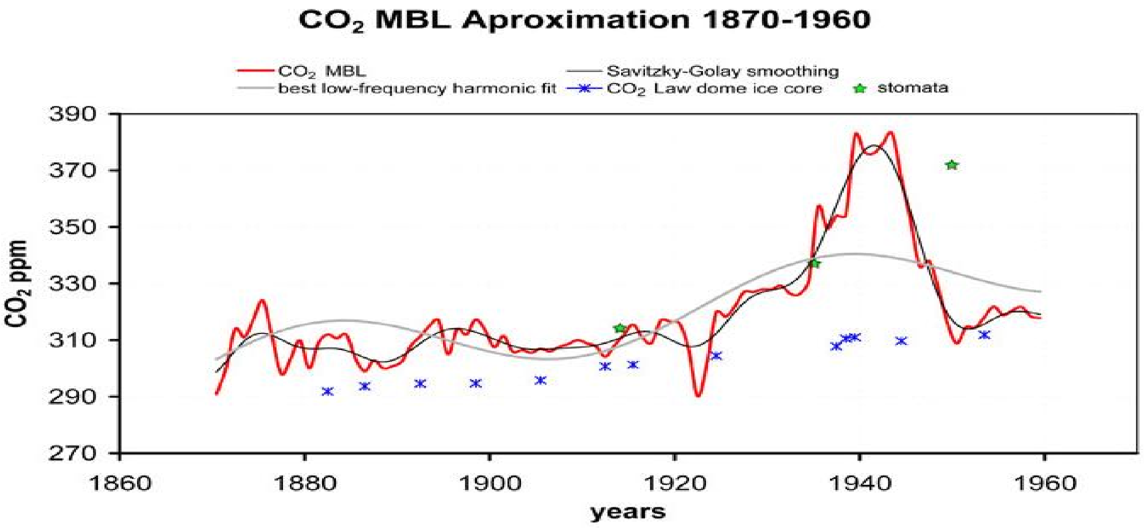


Figure 25. Atmospheric CO_2 background level 1870–1960; black line smoothed by a Savitzky-Golay filter, parameters (Autosignal): width of the moving window: 7, order: 2; passes: 3; grey line= estimated best low-frequency harmonic fit (55.17 years = 18.39 years x 3); blue stars= CO_2 law dome ice core (Etheridge et al. 1998); green stars= stomata Tsuga spec., Jay Bath, Mount Rainier Washington, USA; (Kouwenberg 2005). 1922: phase reversal.

Figure 25 confirms the early analysis of Callendar (1940) that CO_2-levels in the atmosphere have grown since the late 19th century.

The minimum is 289.4 ppm in 1922, the maximum is 383 ppm in 1943, mean = 324.63 and the standard deviation = 19.63 for the period 1870–1960

The average in the 19th century using the reconstructed MBL (1826–1900) is 322.67 ppm. The average in 20th century using a combination of historical and modern data (NOAA) is 331.38 ppm. This suggests a rise of 8.71 ppm or 2.6 % which is well within observed variability and error range of the methods. Data prior to 1826 are not included because of the great uncertainty. If the estimated higher CO_2-levels had been real at the beginning of the 19th century, there would be no difference in the average CO_2-levels between the last two centuries.

Since 1870 two periods of about 60–70 years (1870–1940) and 1949–2009 can be observed with such a slow rise of the CO_2-levels to a maximum level of about +70 ppm in about 1943 and today. The overall rising CO_2-levels since 1870 to today has also been supported by the reconstructed ice core data despite of its much worse resolution compared to the presented data (see Figure 25). The maxima around 1860 and 1940 are in contrast to the published literature. They can be described by an about 70–80 year cycle.

Stomata are tiny openings in the epidermis of leafs to allow gas exchange of the plants. At high atmospheric CO_2-levels leaves adapt by decreasing their density. At low levels they increase their density to absorb more CO_2. The inverse relation is confirmed over different time scales (Royer 2003; van Hoof 2006, 2008). Several reconstructions of atmospheric CO_2-levels exist in modern times. Wagner (2005) reported of low density of stomata found in several species from the swamps of Florida around 1940–50 and since around 1980–2000. The same low stomata density today as in the first half of the 20th century can be seen in Quercus spec. leaves from the Netherlands (van Hoof et al. 2006). CO_2 reconstruction from stomata at Jay Bath, Mount Rainier, Washington, USA (Kouwenberg 2005) in 20th century, show 332 ppm in 1914, 337 ppm in 1935 and 372 ppm in 1950 which fit very well to the CO_2 MBL from direct measurements (Figure 25).

These stomata data confirm the CO_2 MBL reconstruction as well as the raw data showing high CO_2-levels in the 1930s and 40s at higher temperatures. This is the pre-condition for the inverse stomata/CO_2 relation.

If we assume the actual CO_2-levels are near a peak, the levels are expected to drop within the next years. The application of spectrum analysis on the historical data set would give more answers.

4. Conclusion

In this paper a new dataset of directly measured CO_2 samples from 1826 is introduced. They have been sampled and analysed mostly near ground by well known chemical methods (Pettenkofer process and related volumetric methods) by experienced scientists since the early 19th century (see station and author list in the supplemental data) which contributed to the evolution of gas analytical methods. From an estimated total sum of more than 200 000 single samples collected since 1800 in the northern and southern hemispheres, the author has selected 97 404 samples from 901 stations compiled in 87 data files (supplemental data). A thorough literature review revealed that these data are only partly known to climate science and had been rejected and ignored without a thorough validation of the quality, sampling and analysing methods behind. This new dataset contains high quality data including vertical profiles which can be easily compared to today´s standards. The selection process was characterized by using only data sampled by known methods and from locations which allow a validation of local influences and air masses.

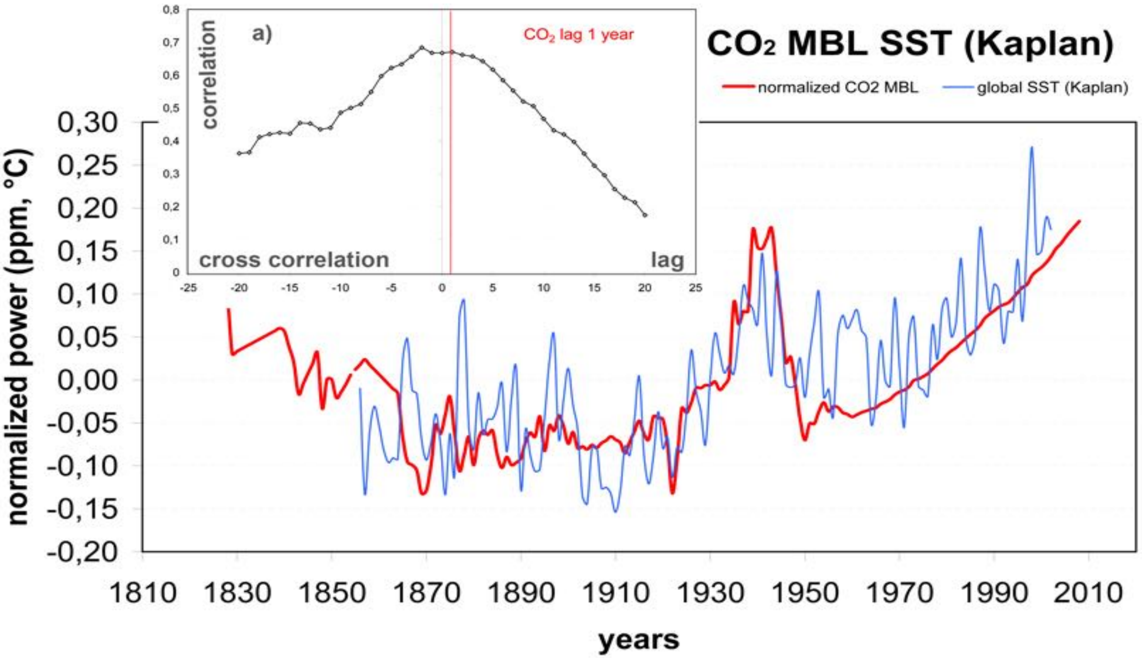


Figure 26. Annual atmospheric CO_2 background level 1856–2008 compared to SST (Kaplan, KNMI); red line: CO_2 MBL reconstruction 1826–1959 (Beck), 1960–2008 (MLO); blue line: Annual SST (Kaplan) 1856 –2003; a) cross correlation of SST and CO_2 MBL showing correlation of r=0.668 and a lag of 1 year for CO_2 after global SST.

To make these near ground data comparable to modern CO_2 background data compiled under international standards, new methods had to be developed to estimate the marine boundary level (MBL) or background levels in the higher troposphere from near ground data. Using modern vertical profiles available from the WMO and NOAA sampling networks, the author was able to establish 4 estimation methods which allow calculation of the MBL from near ground air mass characteristics. By simple nonlinear regression methods it was possible to estimate the MBL within 1–3 % error range from a comparison of CO_2-levels at higher wind speeds and precipitation levels. The new MBL estimation methods from near ground data are named CWBA, CPBA, CSMBA, CDMBA. They also include a calculation from the daily minimum CO_2-levels initially speculated by C. Keeling (1958)(CDMBA) and an estimate based on the summer minimum levels (CSMBA). These methods have been applied to the 87 historical data series to calculate the historical MBL since 1826 as annual averages within an overall error range of about ±3 % since 1870 (table 11). Missing years are filled by interpolated values. The resolution of these annual new data series is much better compared to reconstructions from ice core and stomata data. Showing 317.8 ppm in 1959, it fits perfect to the modern values of Mauna Loa based CO_2-levels analysed by NDIR sensors despite a calibration against the old methods is not reported by C. Keeling.

The most remarkable conclusion from the contour of the new CO_2 MBL data is that CO_2-levels between 1930 and 1950 have been as high as today. Furthermore, a distinct dip can be observed around 1922.

The data also suggest higher levels in the first half of the 19th century than reconstructed from commonly used ice cores. Using modern MLO CO_2 data, we can calculate a centennial average for the 20th century 1901–2000 of 331.38 ppm and of a MBL in the 19th century (1826–1900) of 322.67. This is a growth rate of +2.6 % in contrast to about 30 % as derived from ice cores and therefore within measurement variability. Analysing the new series of directly measured CO_2 MBL levels from 1926 to 2010 suggests a possible cyclic behaviour.

The CO_2 MBL levels since 1826 to 2008 show a good correlation to the global SST (Kaplan, KNMI; see Figure 26) with a CO_2 lag of 1 year after SST from cross correlation (Figure 26a). Kuo et al. (1990) had derived 5 months lag from MLO data alone.

Table 12. Summary of acronyms and abbreviations

Ameriflux	The AmeriFlux network was established in 1996. The network provides continuous observations of ecosystem level exchanges of CO_2, water, energy and momentum spanning diurnal, synoptic, seasonal, and interannual time scales and is currently composed of sites from North America, Central America, and South America
BASF	Badische Anilin und Soda Fabrik
C	Celsius
CDMBA	CO_2 daily minimum background approximation
CGER	Center for Global Environmental Research
COADS	Comprehensive Ocean-Atmosphere Data Set
CPBA	CO_2 precipitation background approximation
CSMBA	CO_2 summer minimum background approximation,
CWBA	CO_2 wind speed background approximation
E	east
GlobalView-CO2	is a product of the Cooperative Atmospheric Data Integration Project. The project is coordinated and maintained by the Carbon Cycle Greenhouse Gases Group of the National Oceanic and Atmospheric Administration, Earth System Research Laboratory (NOAA ESRL),
IPCC	Intergovernmental Panel on Climate Change
KNMI	Koninklijk Nederlands Meteorologisch Instituut
KSLA	Kungl. Skogs- och Lantbruksakademien, (Royal Swedish Academy of Agriculture and Forestry)
m	meter
MBL	marine boundary layer
mg	milli gram
MLO	Mauna Loa Observatory, Hawaii
N	north
NCDC	National Climatic Data Center
NDIR	nondispersive infrared sensor
NOAA	National Oceanic and Atmospheric Administration
ppm	parts per million (volume)
ppmv	parts per million per volume
prec	precipitation
RICO C	Riedel gas analyser type C
S	south
SEAS	seasonal cycle
Sm	summer minimum
SST	sea surface temperature
URAS	Ultrarot-Absorptionsschreiber (infrared absortion writer),
W	west
WDCGG	World Data Centre for Greenhouse Gases
WMO	World Meteorological Organisation
ws	wind speed

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3) CO2_Literature_1800-1960.pdf

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5) Samplesused-jan2010.xls

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CO_2 1800-1960 Historical References, Chemical Methods

Links to digitized sources

Note: Montsouris 1877-1910

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CO_2 Sampling Stations - Chemical Methods 1800-1960

Note: Montsouris 1877-1910; all stations listed with their first year

year	Station	Method
	97 409 samples used at 901 stations	
1809	•**Investigator:** Theodore de Saussure, Prof. mineralogy, geology University Geneva (Switzerland) •**Location:** Meadow near the village of Chambesy near Geneva (Switzerland), slightly inclined, 250 m from the Lake of Geneva, well ventilated, soil: clay, , rural area; •Lat 46,15N. Lon: 6,8 E. •**Elevation**: 1,3 m above ground, 16 m above sea level of Lake Geneva, 388m masl •Mountains around Chambesy 900 – 1300m height **Sampling time:** 1809-1816, at noon 1826-1830 **Samples:** 225 since 1809, since 1826 205; 8 samples at mountains **Meteorolog. Parameters:** daytime, precipitation, weather Station 1, 2	•**Gravimetric determination** of Barium carbonate (sulphate) •**Sampling:** Simultaneous measurements day/night -city/rural area-water/land, vegetation, mountains, rain, winter, summer; 6 times sampling in one location, 225 single samples since 1826 •**Duration of analysis:** 24 hours. •**Methodical details:** 35-45 Litre round flask with fixed lock, stop cocks and seals, air pumping by air pump; measurement of temperature, air pressure and humidity; 100 g baryta water, absorption time 1 h. (60-80 x shaking/minute); Barium carbonate in baryta water was separated after absorption to wash flask; Elution with HCl several times rest of baryta in flask (1mass unit HCl on 15 mass units water = about. 0,8M), joining precipitations and precipitation with Na_2SO_4.;Sufficient absorption time of solutions (-24 hours). Resulting dry barium carbonate was weighed (mg scales) and mass of bound CO_2 was calculated. **Remarks and Errors:** •Results relatively stable on a high level compared to later series 1830-1850; minimum 0,.2 mg weighing error because of variable mass of absorption equipment [Warburg 1905; Hlasiwetz 1856 p.9]; 3rd procedure according to Thenard: weight of $Ba(CO_3)$: 0,966g ; 391 ppm CO_2; min. ~21% error, seals in leather is a cause of erroneous high values after Müntz (1880); max. 2,5% error by hygroscopic glass walls; resulted weights of CO2 by using around 30 l is in the order of about 30 mg using 330 ppm. Weight errors of glass apparatures are in then same error range up to 32 mg [Hlasiwetz 1856] **Estimated error:** 20%
1839	**Investigator: Jean-Baptiste Boussingault**, Prof. chemistry Lyon, Paris **Location:** Paris (48N 2E), periphery, Alsace Pechelbronn (Alsace, France) lat 48,93N, 7,83E Liebfrauenberg (Alsace) lat 48,23N 7,38E Andilly lat 49N 2,3E, rural area **Elevation:** 1-2 m Paris: 129 masl Pechelbronn: masl 153-199 m Liebfrauenberg: masl 222m Andilly: masl 278 **Sampling time:** 1839-1843; at Paris 1839-1841 during 142 days In Alsace Nov.- 1839-March 1840 during 49 days; at Andilly and Paris, College de France 1843 Sept—	• **Gravimetric determination** of Sodium sulfate Citation from Hlasiwetz 1856: Method after Carl Emmanuel Brunner, Professor of chemistry in Bern (Switzerland) •About 25 Litre air flows through tubes with H_2SO_4 on pumice, then through tubes with KOH, then again through tubes with H2SO4. Amount of CO_2 was calculated by rise of mass in the tubes with KOH and H2SO4. Citation from Brunner, M. ´Determination de l´acide carbonique´; Annales de Chimie, 3, 1841, p. 312 **Remarks and errors:** •U-tubes containing H_2SO_4 absorb CO_2, therefore resulting in too low values (~20 ppm according to Spring 1885, 7-10% according to Hlasiwetz

	Nov. **Samples:** Alsace 19, Paris 190, Paris/Montmorency 1843: 22; total: 231 **Meteorolog. Parameters:** daytime, precipitation, weather	1856); Amount of CO_2 too small for accurate weighing; min. 0,2mg error [Warburg 1902]; max. 2,5% error by hygroscopic glass walls; resulted weights of CO_2 by using around 30 l is in the order of about 30 mg using 330 ppm. Weight errors of glass apparatures are in then same error range up to 32 mg [Hlasiwetz 1856]; large fluctuation of values (-100%) shown by most series done by several authors **Estimated error:** 50%
1847	**Investigator: Bernhard Carl Lewy**, (Danmark), Prof. chemistry, Bogota **Location:** France, Atlant. Ocean, Caribbean, Colombia, rural area Sept. 1847 at Paris (48N 2E), Nov./Dec. 1847 Le Harvre (49N, 0,1E) Atlantic Ocean Dec. 1847: 47,3N, -8,5E, 47N, -11E, 33,4N, -18,35E, 21,45N, -39,3E, 21,9N, -40,25E, 20,35N, -41,35E, 15,49N, -62,48E, 14,6N, -68,4E, 12,5N, -76E Colombia: Rio Magdalena San Bernardo 4,4N, -74,2, Ambalema 4,78N, -76,7E, Esperanza 7,39N, -73,37E, Santa Ana 7,14N, -73,29E, Honda 7,28N, -73,25E, Guaduas, 5,7N, -74,5E, Bogota 4N, **Elevation:** Paris, 30 masl, Le Havre 8 masl, Rio Magdalena 38 masl, Ambalema 282 masl, Esperanza 396 masl, Honda 242 masl, Guaduas 996 masl, Santa Ana 998 masl, Bogota 2645 masl **Sampling time:** Sept 1847-Aug 1848, High amount of volcanoes in the Rio Magdalena area, high CO2 low O2 **Samples:** Paris 5, Le Havre: 4, Atlantic Ocean: 11, Colombia: 11; Bogota 14: total: 45 **Meteorolog. Parameters:** daytime, temperature, pressure, precipitation, weather	**Volumetric determination** according to Regnault/Reiset Part of samples in flasks are analysed 18-20 months after sampling at Paris •Hydrogen Eudiometer; air flow trough tubes with pumice and KOH, then baryta water. Adding H_2SO_4 the potassium carbonate is transformed to potassium sulphate liberating CO_2, after that measuring change of volume. • **Remarks and errors:** Open system using long connections between CO_2 absorption and Eudiometer, partly consisting of caoutchouk; no control of heat of absorption temperature (approx. 20,000 cal/g Mol; Schuftan 1933): Temperature change gas of 1° = change of vol/pressure by 0.34 %; [Schuftan 1933]. Further methodical errors by absorption of CO_2 in H_2SO_4 resulting in too low values (about –20 ppm). (Using H_2SO_4 for drying air see Regnault/Reiset, Annales de chimie, 24, 1871 p.258 [Regnault 1871]) Standard dev. of samples from Columbia= 641,3
1848	**Investigator: Richard F. Marchand**, Prof. chemistry at Halle (Germany) Conditions not researchable **Sampling time** 1848-1850 **Samples:** 150 No further details researchable	**Gravimetric method similar Brunner** Several samples showing very high and low levels **Estimated error:** >20-50%
1851	**Investigator: Ch. Mène** **Location:** Paris (48N 2E), **Sampling time:** Aug 1851 **Samples:** 12 **Meteorolog. Parameters:** daytime, temperature, No further details researchable **Investigator: Adolf Schlagintweit**, Prof. geography Munich (Germany) **Location:** Swiss/Italian Alps (45N, 7,51E), rural area **Elevation:** 1370-1862 masl **Sampling time:** Sept 1851 **Samples:** 16 (Alps+ Berlin) **Meteorolog. Parameters:** daytime, temperature, pressure, weather	**Mène:** Method according to Boussingault and titrimetric **Remarks and errors:** Drying air by sulphuric acid, -20 ppm error according to Spring and Hlasiwetz **Estimated error:** selected data >20 % No further details researchable **Schlagintweit:** gravimetric; passing air through tubes of sulphuric acid or $CaCl_2$ solution for drying; absorption in 3 tubes with KOH;. Weighing the absorbed solution; **Remarks and errors:** Drying air by sulphuric acid, -20 ppm error according to Spring and Hlasiwetz; weighing errors, gravimetric see Brunner (Boussingault. **Estimated error:** selected data >20 %

1854	**Investigator: August Vogel jun.**, Prof. organic chemistry Munich (Germany) **Location:** Munich (48N, 11,35E), city free place **Elevation:** 519 masl **Sampling time:** Aug. 1854 **Samples:** 11 **Meteorolog. Parameters:** -	Gravimetric after Brunner see above **Remarks and errors:** Drying air by sulphuric acid, -20 ppm error according to Spring and Hlasiwetz; weighing errors, gravimetric see Brunner (Boussingault. **Estimated error:** selected data about 20 % using selected data
1856	**Investigator: Hugo v. Gilm.**, Prof. chemistry Innsbruck (Austria) **Location:** Innsbruck (Austria) (47N, 11,39E) garden of the university **Elevation:** 573 masl **Sampling time:** Nov. 1856- March 1857 **Samples:** 19 **Meteorolog. Parameters:** temp. pressure, state of the atmosphere Station 16	**Gravimetric/volumetric**, Aspiration of approx. 60 L air and absorption in baryta water; dilution of filtrated Barium carbonate in HCl, drying of produced Barium chloride. Subsequent determination of Chlorine by titration after Mohr; amount Chlorine = CO_2. Calibration: Determination of a weighed amount of $BaCO_3$ in a CO_2 free flask. Decomposition of $BaCO_3$ by H_2SO_4, measurement of deliberated amount of $CO_{2,}$ result: **Remarks and errors:** weighing errors, gravimetric see Brunner/ Boussingault. **Estimated error:** 10%; about 2,8 % using selected data
1863	**Investigator: Franz Schulze.**, Prof. chemistry Rostock (Germany) **Location:** western balcony of university of Rostock (54N, 12,06E) , free ventilation **Elevation:** about 19 masl **Sampling time:** Oct. 1863- Dec 1864 **Samples:** 431 **Meteorolog. Parameters:** daytime, temp. pressure, Wind direction, precipitation, state of the atmosphere Station 18	**Pettenkofer titrimetric** •Flask method/tube method; approx. 25 Litre air per analysis by using 2 l flasks/tubes; baryta water for absorption of CO_2 and oxalic acid for titration of $CaCO_3$; indicator for showing endpoint of titration; about 30 minutes per analysis. **Remarks and errors:** Independent on temperature, easy design, fast speed, (30 min) high accuracy possible (1-3% Kauko 1935), errors possible by sampling air, contamination of baryta water during titration and too small air volume for testing. Schulze had used max. 25 l air for one analysis . **Estimated error:** +--10%

1864	**Investigator: Agnus Smith.**, Dr. chemistry Manchester (UK) **Location:** various places Scotland, (56,1-3N, -3,23E) rural areas, hills 1 **Elevation:** 250-1400 masl **Sampling time:** 1864-1865 **Samples**: 200 London/Manchester, 158 Scotland total: 358 **Meteorolog. Parameters:** daytime, temp. pressure, wind, state of the atmosphere Station-57	**Pettenkofer flask method** as Schulze Baryta water, Oxalic acid, titration **Estimated error:** +--10%
1865	**Investigator: T.E Thorpe**, Prof. chemistry Leeds (UK) **Location:** along the coasts, (54,21N, -2,11E) Irish sea **Elevation:** 2 masl **Sampling time:** summer 1865-summer 1866 **Samples:** 77 Irish channel; 31 in tropical Brasil (Letts&Blake) **Meteorolog. Parameters:** daytime, temp. pressure, wind direction, speed, state of the atmosphere	**Pettenkofer flask method** as Schulze 5l flasks,modified Baryta water, Oxalic acid, titration **Estimated error:** +--3%

	Station 157	
1868	**Investigator: Franz Schulze.**, Prof. chemistry Rostock (Germany) **Location:** university of Rostock (54N, 12,06E), direction Blücherplatz, periphery of the city, free ventilation **Elevation:** about 19 masl **Sampling time:** Oct. 1868- July 1871, 2 samples daily **Samples:** 1600 **Meteorolog. Parameters:** daytime, temp. pressure, wind direction, precipitation, state of the atmosphere station 158	**Pettenkofer flask method** , titrimetric, modified 25 l flasks, 66 l Aspirator Baryta water, Oxalic acid, titration **Estimated error:** +--3%
1872	**Investigator: J.A Reiset.**, Prof. chemistry Paris **Location:** 8m from Dieppe (France) (49N, 1,07E), agricultural station, rural area, free ventilation **Elevation:** about 96 masl **Sampling time:** Sept. 1872 – Oct. 1873, **Samples:** 92 field, 27 young forest, 14 clover/alfalfa **Meteorolog. Parameters:** daytime, temp. pressure, wind direction, precipitation, state of the atmosphere station 161 **Investigator: Eugène Risler.**, Prof. agronomie, Paris **Location:** Calèves (Nyon, Switzerland) (46N, 6,63E), rural area, free ventilation **Elevation:** 420 masl **Sampling time:** Aug. 1872 – July 1873 **Samples:** 365 **Meteorolog. Parameters:** not known Station 162 **Investigator: Wilhelm Henneberg.**, Prof. chemistry Göttingen (Germany) **Location:** Weende (Germany) (51,5N, 9,95E), agricultural station, rural area **Elevation:** about 237 masl **Sampling time:** May - July 1872, **Samples: >**17 **Meteorolog. Parameters:** temperature Station 163	**Reiset:** **Pettenkofer variant** mobile analyser, 600 l aspirator (Reiset´s tower); drying by sulphuric acid, absorption in barium hydroxide; titration by resulting carbonate by sulphuric acid. **Remarks and errors:** Sulphuric acid error of about 20-30 ppm according to Hlasiwetz [1848] and Spring (1883); Bunsen absorption coefficient H_2SO_4 at 25°C = 0,96; H_2O at 25°C=0,759 [IUPAC NIST Solubility database] **Estimated error:** correction by +20 ppm; error+--3% **Risler:** **Pettenkofer variant** no more details researchable **Henneberg, Weende:** **Pettenkofer flask method** as Schulze 5l flasks,modified Baryta water, Oxalic acid, titration **Estimated error:** +--3%
1873	**Investigator: Pierre Truchot.**, Prof. chemistry, Clermont-Ferrand **Location:** some kms from Clermont-Ferrand, (45N, 1,05E), rural area, free ventilation **Elevation:** near Clermont Ferrand 395 masl; **Sampling time:** July-Aug. 1873, day/night **Samples:** 49 **Meteorolog. Parameters:** temp., pressure Station 164	**Pettenkofer method** 10-20l air absorbed in baryta water, titration the carbonate with sulphuric acid **Estimated error:** +--3-10%

1874	**Investigator: Franz Farsky.**, Dr. chemistry, director of the agricultural station at Tabor, Czech Republic **Location:** agricultural station outside Tabor, (49,25N, 14,6E), rural area, free ventilation, 4,48m above ground NNW **Elevation:** 423 masl; **Sampling time:** Oct. 1874 -Aug. 1875 Spring /summer afternoon, autumn/winter at noon Station165 **Samples**: 295 **Meteorolog. Parameters:** temp., pressure, wind direction, precipitation, weather	**Pettenkofer variant of Fittbogen** Air passing from outside (4,48m elevation) NNW in $CaCl_2$ tubes (drying) and then in a absorption tube with baryta water then in a Brunner 30 l aspirator. Absorbing time 4 h. Titration was done with hydrochloric acid or oxalic acid. **Estimated error:** +--3%
	Investigator: P. Hässelbarth, Dr. chemistry, **J. Fittbogen** Prof. chemistry, director agricultural station Dahme **Location:** Dahme (Prussia) lat 51N, 11,04 lon; agricultural station, rural area **Elevation:** 87 masl, sampling height: 2,85m **Sampling time:** Sept. 1874-Aug 1875 **Samples**: 347 **Meteorolog. Parameters:** temp., pressure, wind direction, precipitation, weather Station 166	**Pettenkofer variant of Fittbogen** Air passing from eastern, outside of laboratory (2,85m elevation) in $CaCl_2$ tubes (drying) and then in a absorption tube with baryta water then in a 32 l aspirator. Absorbing time 5 h. Titration was done with oxalic acid. **Estimated error:** +--3%
1875	**Investigator: Gaston Tissandier**, **Location:** Paris : 890 masl 1000 masl during balloon flight (Zenith) **Elevation:** 87 masl, sampling height: 2,85m **Sampling time:** March. 1874 **Samples**: 2 **Meteorolog. Parameters:** temp., pressure, wind direction, weather Station 167	**Volumetric determination** according to Regnault/Reiset •air flow trough tubes with pumice and KOH, then baryta water. 22 l Aspirator. Adding H_2SO_4 the potassium carbonate is transformed to potassium sulphate liberating CO_2, after that measuring change of volume. Using 66 l air **Remarks and errors:** Open system using long connections between CO_2 absorption and Eudiometer, partly consisting of caoutchouc; no control of heat of absorption temperature (approx. 20,000 cal/g Mol; Schuftan 1933): Temperature change gas of 1° = change of vol/pressure by 0.34 %; [Schuftan 1933]. Further methodical errors by absorption of CO_2 in H_2SO_4 resulting in too low values (about –20 ppm). **Estimated error:** +--8%
	Investigator: Peter Claesson, Dr. chemist **Location:** Lund (Sweden) lat 55, 13,12E lon **Elevation:** 47 masl **Sampling time:** Nov/Dec. 1875 **Samples**: 31 **Meteorolog. Parameters:** - Station 168	**Claesson** **Pettenkofer method** 50 air passed through CaCl2, absorbed in baryta water, titration the carbonate **Estimated error:** +--3-10%
1876	**Investigator: E.L. Moss**, Dr. physician **Location:** Arctic, Greenland lat 82,7, -20E lon **Elevation:** - **Sampling time:** Dec 1875, Jan/Feb. 1876 **Samples**: 3 **Meteorolog. Parameters:** temp., pressure, weather Station 169	**Pettenkofer method** 47, 95, 19 l samples of air absorbed in baryta water, titration the carbonate **Estimated error:** +--3-10%

1877	**Investigator: Hippolyte Marié-Davy**, Prof. physicist, Montpellier; **Albert Lévy**, Dr. physiciste **Location:** Paris 48,58N, 2,27E Montsouris observatory **Elevation:** 75 masl **Sampling time:** daily 1877-1912, noon **Samples**: 12 000 **Meteorolog. Parameters:** temp., pressure, wind, precipitation, weather Station 170	**Pettenkofer method** Absorption of dry air in baryta water /sodium hydroxide, titration with HCl (since 1892 H_2SO_4) Changing method in July 1890 leads to a shift of + 18,5 ppm in SEAS **Remarks and errors:** First years 1877-July 1890 values too low, corrected by +18,5 **Estimated error:** +--3%
1879	**Investigator: J.A Reiset.**, Prof. chemistry Paris **Location:** 8km from Dieppe (France) (49N, 1,07E), agricultural station, rural area, free ventilation, the North Sea in NE direction **Elevation:** about 96 masl +4m sampling height **Sampling time:** June – Nov. 1879, day/night, June-Aug. 1880 **Samples:** 91 (1979), 37 (1880), **Meteorolog. Parameters:** daytime, temp. pressure, wind direction, precipitation, state of the atmosphere station 171	**Reiset:** **Pettenkofer and volumetric variant** according to Regnault mobile analyser, 600 l aspirator (Reiset´s tower); , drying by sulphuric acid, absorption in barium hydroxide (12 hours); titration the resulting carbonate by sulphuric acid. Furthermore volumetric determination. **Remarks and errors:** Sulphuric acid error of about 20-30 ppm according to Hlasiwetz [1848] and Spring (1883); Bunsen absorption coefficient H_2SO_4 at 25°C = 0,96; H_2O at 25°C=0,759 [IUPAC NIST Solubility database] **Estimated error:** correction by +20 ppm; error+--3%
	Investigator: George F. Armstrong, Prof. engineering, Leeds (UK) **Location:** village of Grasmere 54,45N, -3,0E , rural area at Lake Grasmere surrounded by hills, meadow **Elevation:** 61 masl +1,50 sampling height **Sampling time:** July - Oct. 1879, day/night **Samples**: 115 (53 day, 62 night) **Meteorolog. Parameters:** temp., pressure, wind, precipitation, weather Station 172	**Armstrong:** **Pettenkofer variant** 4 x 10l jars; air passed through Baryta water titrated with sulphuric acid, quadruple sampling
	Investigator: Ippolito Macagno, Prof., chemistry, director of the royal agricultural station of Palermo (Silcilia)) **Location:** Palermo, astronomical observatory 38,6N, 13,21E , **Elevation:** 72 masl **Sampling time:** Feb - Aug. 1879 **Samples**: 21 (3 per month) **Meteorolog. Parameters:** temp., pressure, O2, precipitation Station 173	**Macagno:** Assumption**: Pettenkofer variant** data measured in 100 l air **Estimated error:** -3-10%
1881	**Investigator: Achille Müntz**, Prof., chemistry, National Institute of Agronomy Paris **Location:** Paris, 48N, 2,14E, Plaines de Vincennes (rural area) 48,84N, 2,44E; Pic Midi 38,6N, 13,21E , **Elevation:** Paris:129 masl , Pleines des Vincennes 52 masl, Pic Midi: 2877 masl **Sampling time:** Dec. 1880. - Dec. 1881 **Samples**: Paris: 42, Plaine d. V.: 35, Pic Midi: 14 **Meteorolog. Parameters:** temp., pressure, weather, precipitation	**Volumetric after Regnault** 300l air dried by sulphuric acid was absorbed in KOH; addition of sulphuric acid set free CO2, volume measured by manometer. **Remarks and errors:** Open system, no means of controlling heat of absorption; sulphuric acid absorbed parts of CO2, error of about 20-30 ppm according to Hlasiwetz [1848] and Spring (1883); Bunsen absorption coefficient H_2SO_4 at 25°C = 0,96; H_2O at

	Station 174-176	25°C=0,759 [IUPAC NIST Solubility database] **Estimated error:** correction : + 20 ppm , after correction: +--3%
1882	**Investigator: Achille Müntz**, Prof., chemistry, National Institute of Agronomy Paris **Location:** Venus expedition: Haiti 18N, -72E, Florida 29,9N, -81E, Martinique 14N, -61E, Mexico 26N, -111E, Santa Cruz 34,35S, -71,74E, Chubut 43,18S, -65E, Cerro Negro 36,9S, -72,5 E **Elevation:** Haiti 355 masl , Florida 4 masl, Martinique 1masl , Mexico, Santa Cruz 520 masl, Chubut 50 masl, Cerro-Negro 153 masl **Sampling time:** Haiti Nov. 1882. - Jan. 1883; Florida Nov-Dec. 1882, Martinique Oct. –Dec. 1882, Mexico Nov.-Dec. 1882, Santa Cruz Patagonia Oct-Dec. 1882, Chubut Nov.-Dec 1882,, Cerro-Negro Dec 1882 **Samples**: Haiti: 8, Florida: 7, Martinique: 5, Mexico 3, Santa Cruz 10, Chubut 2, Cerro Negro (Chile) 5; total 38 (1882); 2 at Haiti 1883 **Meteorolog. Parameters:** temp., pressure, wind, weather, precipitation Station 177-183 The French expedition to Cap Horn Sept 1882-1883 **Investigator:** Sampling by **Dr. Paul Hyades**, physician of the expedition; A. Müntz, Prof. chemistry, Paris **Location:** Orange Bay Hoste Island, 55,3N, -68,05E **Elevation:** 6 masl **Sampling time:** Sept. 1882-Sept. 1883 **Samples**: 6 (1882), 39 , + 6 (=44 in 1883) on the ship in south Atlantic, 1 islands of Cape Verde **Meteorolog. Parameters:** temp., pressure, wind direction, precipitation, weather Station184, 185	**Volumetric after Regnault** Trained persons on the ships of the expedition sampled air by passing it through prepared flasks filled with KOH. They had been analysed back in Paris by a modified apparature since 1881 with a 160 l aspirator, **Remarks and errors:** Sulphuric acid error, values too low; correction + 20 pp (according to Spring 1883). Blocked upwelling, by ice, therefore too low values, not used (Poisson et al. 1987, Deep Sea Research, Vol 34, no. 7, p. 1255); possible absorption or decomposing error in glass vessels stored for max. 1 year by reaction to potassium silicate, changing solution equilibrium and freeing CO2. **Estimated error:** correction : + 20 ppm , after correction: +--3% **French expedition to Cap Horn** Air was sampled in glass flasks filled with KOH, locked and stored in a metal closing. Analysis was done later in 1883 in Paris using the Müntz volumetric gasometer (see above). **Remarks and errors:** See above
1883	**Investigator: Walthère Spring** et al., Prof., mine engineering, chemistry, university of Liege (Belgium) **Location:** Chemical institute at Liege, 50,3N, 5,5E , direction the river of Meuse **Elevation:** 63 masl + 5 m sampling height **Sampling time:** Feb. 1883 –Feb. 1884 **Samples**: 266 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 186 **E. Ebermayer see 1884** **Location:** Tölz, Forsthaus 47,8N, 12,5E, **Samples:** june 1883 - December: 29	**Pettenkofer variant** Passing air through baryta water and an aspirator of 114,6 l, titration by hydrochloric acid without drying. Checking for optimal passage, absorption speed of air stream, absorption of CO2 by cautschouc, and absorption if using sulphuric acid for drying. **Remarks and Errors:** Calibrated apperature and carefully controlled conditions, enhanced CO2 level in the city by winds from the industrial centres, enhanced levels by carbon oxidation from coal in the soil **Estimated error:** +--2-3%
1884	**Investigator: Ernst Ebermayer**, Prof. agricultural chemistry, university of Munich **Location:** Bavarian forests, 47,6N, 12,5E , **Elevation:** 813 masl + 1,5 m sampling hight **Sampling time:** Oct. 1883 –Nov. 1884 **Samples**: 68 (40 in 1884) **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather	**Pettenkofer method** **Estimated error:** +- 3%

	Station 187 **Investigator: Walther Hempel**, Prof. chemistry, university of Dresden **Location:** Dresden, 51,3N, 13,7E , **Elevation:** 111 masl + 15 m sampling height **Sampling time:** Oct. 1884 –Dec. 1884 daily **Samples**: 63 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 188	**Hempel** Volumetric Constant volume, constant temperature 15°C, variable pressure, Air from the roof of the laboratory or 100ccm glass flasks; Measurement of gases in H20 satured state In parallel check of 1 l air with Pettenkofer-Hesse method **Estimated error:** +--3%
1885	**Investigator: William Marcet**, Dr. pysician, Edinburgh/London **Location:** Geneve, 46,3N, 1E , **Elevation:** 388 masl + mountains **Sampling time:** Aug. 1885 –Sept. 1885 **Samples**: 51 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 189 **Investigator: Reinhart Blochmann** Prof. chemistry, University of Koenigsberg **Location:** Koenigsberg /East Prussia, 53,7N, 20,5E **Elevation:** 15 masl **Sampling time:** 1885 **Samples**: 1345 (1200 on land, 44 over sea) Station 703	**Volumetric** Absorption in KOH and volumetric determination of change of air volume after absorption, Heat control by placing the absorbing vessel in temperature controlled bath **Estimated error:** +--2-3% **Blochmann** Pettenkofer variant Air is absorbed in baryta water and titrated **Estimated error:** +- 3%
1886	**Investigator: Thomas van Nuys** Prof. chemistry, University of Indiana **Location:** University park near Bloomington /Indiana, 39,10N, -86,5E **Elevation:** 228 masl + 50 cm **Sampling time:** April 1886 10:00 am **Samples**: 18 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 190 **Investigator: Giorgio Roster** Prof. chemistry, University of Florence **Location:** Florence, 43N, 11,15E **Elevation:** 38 masl + 18 m **Sampling time:** Jan-Dec 1886 **Samples**: 9 Station 191	**Pettenkofer variant** Absorption of air in baryta water **Roster** Pettenkofer variant **Remarks and Errors:** Inverted SEAS (summer high, winter low) **Estimated error:** +--3% in summer
	Investigator: Julius Uffelmann Prof. physician, University of Rostock, director of the institute of hygiene **Location:** Rostock, 54N, 12E court of the university 20 cm above pavement, outside the city, shore of Baltic Sea **Elevation:** 19 masl + 6 m **Sampling time:** Oct 1886 – Sept 1887 daily 11:00 am **Samples**: 92 in 1886; total 420 (26 in rural area) **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 193 **Investigator: Nils Edvard Selander** Dr. physician, Karolinska University Solna (Sweden) **Location:** Stockholm, 59N, 18,3E , fortress vaxholm directly at the sea side	**Uffelmann** **Pettenkofer method** Air passed through 4l glass flasks, shaking for 1minute and leaving flasks for 24 hours before titration with oxalic acid. Checking time before titrating: ½ hour 330 ppm, 8 hours 342 ppm (+3.6 %) **Selander** Not researched, Pettenkofer or volumetric Petteson **Estimated error:** +- 2-3%

	Elevation: 10 masl **Sampling time:** Oct 1887 – June 1888 **Samples**: total 263; 92 in 1886 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 194	
1887	**Investigator: Victor Feldt** Dr. physician, University of Tartu **Location:** Tartu, 55N, 26,7E **Elevation:** 38 masl + 36 m place of the church "Domplatz" **Sampling time:** Feb – May 1887 several times a day **Samples**: 377 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 195 Montsouris see above	**Pettenkofer method** Air stream passed baryta water, , three 6l flasks were filled, shaked for ½ hour, titration by oxalic acid without waiting for a precipitate 20 l aspirator, titration directly in unclear baryta water **Remarks and Errors:** Too low values (- 187 ppm) 16th April 1887) because of small air volume, titration directly in the unclear baryta water without waiting; methodical problems see the same values day and night. Correction +40 ppm **Estimated error:** +--3 after correction
1888	**Investigator: Jacob Heimann** Dr. physician, University of Tartu **Location:** Tartu, 55N, 26,7E **Elevation:** 38 masl + 36 m place of the church "Domplatz" **Sampling time:** June – Sept 1888 several times a day **Samples**: 350 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather	**Pettenkofer method** Air stream passed baryta water, , three 8-10l flasks were filled, shaked for ½ hour, filtration of the precipitation, titration by oxalic acid 20 l aspirator, titration directly in unclear baryta water **Remarks and Errors:** Too low values (- 187 ppm) 16th April 1887) because of small air volume titration directly in the unclear baryta water without waiting; methodical problems see the same values day and night. Correction +40 ppm **Estimated error:** +--3 after correction
	Investigator: Eugen v. Frey Dr. physician, University of Tartu **Location:** Tartu, 55N, 26,7E **Elevation:** 38 masl + 36 m place of the church "Domplatz" **Sampling time:** Oct – Jan 1889 several times a day **Samples**: 556 (104 outside the city at Ratshof rural area) **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 196	**Pettenkofer** as Feldt and Heimann **Remarks and Errors:** Too low values (- 187 ppm) 16th April 1887) because of small air volume titration directly in the unclear baryta water without waiting; methodical problems see the same values day and night. Correction +40 ppm **Estimated error:** +--3 after correction
	Investigator: Dr. F. Nansen /Augusta Palmqvist/. assistant of O. Pettersson teacher in natural science **Location:** travel over the Atlantic to Greenland, 59,48,5N, -3 – 66,8N, -25,2E **Elevation:** 6 masl + 5 m over sea **Sampling time:** May 1888 – June 1888 sea, Sept 1888 Greenland (2300 masl) **Samples**: 35 over sea, 3 over Greenland in 1888, 395 1888 -1890 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 229	**Pettersson volumetric** Flask samples done by F. Nansen on his expedition 1888, analysed by A. Palmqvist in laboratory of O. Pettersson Absorption of CO_2 in KOH, subsequently measurement of volume change of CO_2-free gas by mercury. Absorbing vessels in tempered water bath ; Temperature control of heat of absorption (H_2CO_3 approx. 20 000 cal/gMol; [Schuftan 1933]) much better than older systems (open e.g. Müntz, Regnault); **Remarks and Errors:** result dependent on temperature and pressure after v. Slyke [van Slyke 1932]. **Estimated error:** +- 1-2%

1889	**Investigator: Arthur Petermann** Prof. acricultural chemistry, director of the agricultural station at Gembloux (Belgium) **Location:** near Gembloux, 50,5N, 4,41E agricultural station **Elevation:** 150 masl + 5 m over a meadow **Sampling time:** May 1889 – April 1891 daily from 9 am **Samples**: 525 **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 230	**Pettenkofer variant (Schlösing-Reiset)** Air was passed trough washing flasks (sulphuric acid) and then absorbed by baryta water in a Pettenkofer tube and titrated by oxalic acid. **Remarks and Errors:** Sulphuric acid error of about 20-30 ppm according to Hlasiwetz [1848] and Spring (1883); Bunsen absorption coefficient H_2SO_4 at 25°C = 0,96; H_2O at 25°C=0,759 [IUPAC NIST Solubility database] **Estimated error:** correction by +20 ppm; error+--3%
	Investigator: Augusta Palmqvist/. assistant of O. Pettersson, teacher in natural science **Location:** Stockholm, 59,17,5N, 18,3E **Elevation:** 20 masl + 2 m **Sampling time:** July 1889 – May 1890, Sept 1888 Greenland **Samples**: 77 in 1889 experimental station **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 231	**Pettersson volumetric** See above **Remarks and Errors:** result dependent on temperature and pressure after v. Slyke [van Slyke 1932]. Very accurate **Estimated error:** +- 1- 2% [Johansson 1898, Skand. Arch. Phys. Bd. 8, p. 93]
1890	**Investigator: Augusta Palmqvist**/. assistant of O. Pettersson, teacher in natural science **Location:** Stockholm, 59,17N, 18,3E Tromsö: Husa: 63,48N, 13,11E Skagerak:57,8N, 13,58E Maseskär: 58,5N, 11,05E Kristineberg: 59,17N, 18E **Elevation:** 20 masl + 2 m **Sampling time:** Jan 1890 – May 1890, Tromsö: Feb- March 1890 Husa: Feb- March 1890 Maseskär: Feb- March 1890 Skagerak: Feb 1890 Kristineberg: June –Aug 1890 **Samples**: 120 in 1890 experimental station Tromsö: 23, Husa: 29, Maseskär: 38, Skagerak: 7, Kristineberg: 64; 1888 -1890: 395 samples **Meteorolog. Parameters:** temp., pressure, wind speed/direction, weather Station 236	**Pettersson volumetric** See above **Remarks and Errors:** result dependent on temperature and pressure after v. Slyke [van Slyke 1932]. Very accurate **Estimated error:** +- 1-2%
	Investigator: Arsenius Lebedinzeff, Dr. chemist, University of Odessa **Location:** Odessa, 46,48N, 30,7E **Elevation:** 50 masl **Sampling time:** April-May 1890 **Samples**: 7 **Meteorolog. Parameters:** temp., pressure, weather Station 704	**Pettenkofer variant** Air was passed trough baryta water and titrated by oxalic acid. **Estimated error:** error+--3%
1893	**Investigator:** Salomon August Andrée, engineer **Location:** over Stockholm, 59,17N, 18,3E , **Elevation:** 0-3830 masl (0, 380, 1200, 2370, 3200, 3830) **Sampling time:**. 15th July 1893 **Samples**: 6 **Meteorolog. Parameters:** temp., pressure, wind,	**Volumetric determination** , Pettersson gas analyzer **Remarks and Errors:** see above; first vertical profile by balloon flight **Estimated error:** +- 1-2%

	weather Station 237	
1896	**Investigator: Carleton Williams, Prof.** chemistry, university of Sheffield **Location:** garden 2,4 km west of Sheffield (UK), 53,4N, 1,5E , **Elevation:** 110 masl **Sampling time:**. Dec 1896-April 1897 **Samples**: 142, + 21 in the city of Sheffield **Meteorolog. Parameters:** temp., pressure, wind, weather Station 238	**Pettenkofer variant according to Smith** Air passed in 10l flask and the for absorption in baryta water, filtration of unclear baryta water, titration with oxalic acid **Estimated error:** +--3%
1897	**Investigator: E.A. Letts, Prof.** chemistry, Queens university of Belfast **Location:** 53,55N, -2.4333E, garden of university **Elevation:** 0 masl +2m **Sampling time:**. March July 1887 **Samples**: 46 **Meteorolog. Parameters: :** temp., pressure, wind, weather Station 239	**Pettenkofer variant** 20 l air were absorbed in baryta water and titrated with HCl **Estimated error:** +--3%
1898	**Investigator: Horace Brown, Dr.** chemistry, Jodrell Lab. Kew Gardens London **Location:** Kew Gardens lat 51,5N; 029W **Elevation:** 15 masl + 1, 5 m **Sampling time:**. July 1898 July 1901 **Samples**: 91 (16 in 1898, 18 in 1899, 29 in 1900, 28 in 1901) **Meteorolog. Parameters: :** weather Station 240	**Pettenkofer variant according to Reiset** 100- 200 l air was passed trough washing flasks (sulphuric acid) and then absorbed by NaOH and double titrated by sulphuric acid. **Remarks and Errors:** Sulphuric acid error of about 20-30 ppm according to Hlasiwetz [1848] and Spring (1883); Bunsen absorption coefficient H_2SO_4 at 25°C = 0,96; H_2O at 25°C=0,759 [IUPAC NIST Solubility database] **Estimated error:** correction by +20 ppm; error+--3%
1902	**Investigator: August Krogh**, Prof., zoology university of Kopenhagen, Nobel awardist 1920 **Location:** Disko Island and coast of Greenland 69,75N, -53,5E, Kuganguak 70,28N, -53,9E, Ingnagnak, Napasiligsuak 70,3N, -54,48E, Igdlorpait 60,45N, -45,33, Avatarpait 70,08N -54,8E, Nordfjord 69,95N -54,5E, Ivisarkut 69,73N, -54,78E, Mellemfjord 69,75N, -54,6E, Dikofjord 69,4N, -53,9E, Sioranguak 69,4N, -53,9E, Uvivak 63,0N, -41,4E **Elevation:** 0 -670 masl + 1, 5 m **Sampling time:**. July – August 1902 **Samples**: 59 **Meteorolog. Parameters: :** pressure, wind direction/speed, weather Station 251	**Volumetric using a modified Haldane apparatus** air in flasks was connected to an absorption vessel with KOH , the volume change of the system was determined by a capillary manometer, heat control is maintained by placing vessels for analysis in water bath. Double determinations **Remarks and Errors:** Original Haldane analyser was modified to get better accuracy as normal of about +- 8-16,6 % Upwelling of warmer water with high phyto/zooplankton bloom in June/July/August at Disko island explains high CO_2 values. Moss 1875 using the Pettenkofer method received the same high values in Greenland 1875/76 (642, 483, 536 ppm) Winds with highest CO_2 from N,**W**,S Oxygen values about constant, conclusion: CO_2 from water **Estimated error:** modified +--3,3 % or +- 10 ppm (300 ppm; +- 1,6% 600 ppm)
1906	**Investigator: René Legendre, Prof.** biology, director biological station at Concarneau **Location:** Concarneau (Normandie) 47,5N, -3,85E **Elevation:** 15 masl **Sampling time:**. Aug/Sept, 1906, July/Aug. 1907 **Samples**: 12 in 1906, 12 in 1907, (total 24)	**Pettenkofer variant** used at Montsouris Observatory Paris see above **Remarks:** Data sampled in July /Aug.1907 directly from the roof of the laboratory at the front of the Atlantic sea

	Meteorolog. Parameters: : temperature, pressure, wind direction, weather Station 252	**Estimated error:** +--3 %
1907	**Investigator: Jens P. Lindhard, Dr.** physiology, Danish North East Greenland expedition **Location:** Greenland Denmarkshavn 76°46'N, 18°46'W **Elevation:** 11 masl **Sampling time:**. 1907-1908 **Samples**: 23 **Meteorolog. Parameters: :** temperature, pressure, wind direction, weather Station 253	**Haldane volumetric variant** , same as Krogh 1902 **Remarks:** Same upwelling on some days found as Krogh Normal average 350 ppm, 2 days <300 ppm, 5 days >400 ppm, max 620 ppm (cited in Benedict 1912**)** **Estimated error:** modified +--3,3 % or +- 10 ppm (300 ppm; +- 1,6% 600 ppm)
1908	**Investigator: Jean Charcot, Dr.** physician, 2nd French Antarctic Expedition 1908-1910 CO_2 Analysis: A. Müntz, Paris **Location:** Cape Horn-Antarctic coast 58S -70S, -65- -121E **Elevation:** 3-10 masl **Sampling time:**. 1908-1910 **Samples**: 10 (1 in 1908) **Meteorolog. Parameters: :** temperature, pressure, wind direction, weather Station 263	**Volumetric after Müntz** Trained persons on the ships of the expedition sampled air by filling it in prepared flasks (5 l). They had been analysed back in Paris by a modified apparature. The flask air was passed through KOH and the connected to a volumeter which measures the volume of freed CO_2 by adding sulphuric acid. **Remarks and errors:** Sulphuric acid error, values too low ; correction + 20 pp (according to Spring 1883). Blocked upwelling, by ice, therefore too low values, not used (Poisson et al. 1987, Deep Sea Research, Vol 34, no. 7, p. 1255); Not used because of strong ocean absorption near ice shields (see Buch) **Estimated error:** correction : + 20 ppm , after correction: +--3%
1909	**Investigator: Francis G. Benedict, Prof.** physiology, Director Boston Nutrition Laboratory **Location:** Boston, west side of nutrition laboratory 42,3N, -71E, ocean, Pikes Peak **Elevation:** Boston 13 masl +10 m; Pikes Pike 4312 masl **Sampling time:**. 1909-1912 **Samples**: Boston :212, ocean air 43, Pikes Peak 9 (1909: 42 samples, 1910: 248, 1911, 297, 1912: 17) total 604 **Meteorolog. Parameters: :** temperature, pressure, wind direction, weather Station 265	**Pettersson – Sonden volumetric** See above **Remarks and Errors:** result dependent on temperature and pressure after v. Slyke [van Slyke 1932]. Very accurate **Estimated error:** +- 1-2%
1911	**Investigator: James Kendall, Prof. chemistry**, Columbia University New York **Location:** Edinburg 55,5N, -10E, Petrograd 54,38N, 62,42E, Stockholm 59,2N, 18E, Columbia University 42,8N, -74,5E **Elevation:** Petrograd 180, Edinburgh 32, Stockholm 20, New York 9 masl **Sampling time:**. Edinburgh Mar 1911 Petrograd Jan 1913, Nobel institute Stockholm June 1913, Columbia University July 1915 **Samples**: 18 Edinburgh, 10 Petrograd, 8 Stockholm, 24 New York, total: 60 **Meteorolog. Parameters: :** not available Columbia Institut fresh breeze from the Hudson river Station 269	**Pettenkofer method, Walker modification** (Walker, J., Estimation of atmospheric carbon dioxide; Chem. Soc, Trans. 77, 1110 (1900)**)** see Letts and Blake p 219-229 double determinations **Remarks**: No meteorological and station conditions available Lowest value March 1911:356 ppm, near the city Lowest value Jan 1913: 356 ppm, near the city Lowest value June 1913: 321ppm rural area Lowest value July 1915 :326 ppm Winds from Hudson River **Estimated error:** at least +- 3%

	Investigator: August Krogh, Prof., zoology university of Kopenhagen, Nobel awardist 1920 **Location:** Kopenhagen **Elevation:** 6 masl **Sampling time:** April 1911- Jan 1912, **Samples:** 200 **Meteorolog. Parameters:** : pressure, wind direction/ speed, weather	**Volumetric using a modified Haldane apparatus** See 1902
1912	**Investigator: Albert Wigand, Prof.** meteorology, Hamburg **Location:** Bitterfeld (Germany) 51,6N, 12,3E **Elevation:** 3260, 4990, 6350, 9100 masl **Sampling time:**. Aug. 1911-Sept 1912 **Samples**: 4 (CO_2) **Meteorolog. Parameters: :** temperature, pressure, wind, weather Station 270	**Fractionated Condensation (Erdmann 1910)** (Erdmann et al. Berichte der deutschen chemischen Gesellschaft, 43, (1910), p. 1702/1708 Air was sampled during ballon flights in 2 l flasks and analysed in the chemical laboratory of Halle (Germany) **Remarks**: CO2 in upper troposphere much lower than expected, possible problem of the condensation method by Erdmann correction by +15 ppm **Estimated error:** at least +- 5%
1917	**Investigator: August Krogh**, Prof., zoology university of Kopenhagen, Nobel awardist 1920 **Location:** Kopenhagen lat 55,7 N, 12,5E **Elevation:** 6 masl **Sampling time:**. May. 1917 - June 1918, **Samples**: 76 **Meteorolog. Parameters: :** - Station 271	**Volumetric similar Haldane /Pettersson** 3 gas burettes each for $O_2/N_2/CO_2$, one measuring burette for saturing with H2O, reading of volume change before and after absorption of moist air in KOH **Remarks:** Air in Kopenhagen is between 310 -370 ppm 2 very accurate values after calibration: 305, 300 ppm (average: 302,5 ppm) Temperature constancy in laboratory **Error:** $O_2/N_2/CO_2$ = <= 0,001% Standard deviation in double determinations for CO_2 is 0,00025 %; +-2,5 %.
1920	**Investigator: Henrik Lundegardh**, Prof., botany Central institute for agricultural research Stockholm **Location:** Hallands Väderö, lat 55,5N, 12,55E **Elevation:** 8 masl **Sampling time:**. 1920 - 1926, May - Sept **Samples**: > 3000 **Meteorolog. Parameters: :** - temperature, pressure, wind , weather Station 272	**Pettenkofer variant** Automatic absorption of moist air in baryta water, titration by HCl controlled by phenolphthalein. Temperature control; result CO2 concentration in mg/l **Remarks:** Half -automatic apparature Additional volumetric analysis of soil air and air over crop fields **Error:** +-1% or +-0,0003 Vol %
1921	**Investigator: Bruno Schulz**, Prof., hydrography University Hamburg, institute of oceanography **Location:** North Sea, Baltic sea, 55N, 12,4E **Elevation:** 8 masl **Sampling time:**. 1921, July- August 1921 **Samples**: 25 **Meteorolog. Parameters: :** - temperature, most important oceanographic parameters Station 297	**Volumetric apparatus by Krogh** see 1917 **Remarks:** Surface water of Baltic Sea supersatured with CO2 more than North Sea **Error:** +-2,5%
1924	**Investigator: Theodor Meinecke** Jr., Dr.phil., Dr., forestry science, University Hannover,	**Pettersson – Sonden volumetric** See above

	Location: Eberswalde, 52,8N, 13,7E **Elevation5** masl **Sampling time:**. 1924, June- October **Samples**: >334 **Meteorolog. Parameters:** temperature, wind, weather Station 298	**Remarks and Errors:** result dependent on temperature and pressure after v. Slyke [van Slyke 1932]. Very accurate selected data 5-32 m over ground after rain **Estimated error:** +- 1-2%
1925	**Investigator: Erich Reinau**, Prof., ecologic agriculture **Location:** Davos, 46,8N, 9,7E **Elevation:** 2 masl **Sampling time:**. August 1925 **Samples**: 53 **Meteorolog. Parameters: :** - temperature, weather Station 299	**Pettersson – Sonden volumetric** See above **Remarks and Errors:** result dependent on temperature and pressure after v. Slyke [van Slyke 1932]. Very accurate selected data after rain **Estimated error:** +- 1-2%
	Investigator: Hermann Wattenberg, Prof. Dr., chemist, Hydrography **Location:** southern Atlantic ocean 0S-72S, **Elevation:** 0 masl **Sampling time:**. June 1925 – May 1927 **Samples**: >10 000, 312 calculated CO_2 values over sea surface **Meteorolog. Parameters: :** - temperature, weather oceanographic parameters Station 611	**Volumetric apparatus by Krogh modified by Buch** see 1917 Buch, K. Uber die Alkalinität, Wasserstoffionenkonzentration, Kohlensäure und Kohlensäuretension im Wasser der Finland umgebenden Meere, Helsingfors : Societas scientiarum fennica, 1917. **Remarks:** Measured values higher according to Buch **Error:** +-1-2 %
1926	**Investigator: D. Florentin**, Dr., chemist, Laboratoire municipale de Paris **Location:** Paris, Laboratoire de Villejuif, 48N, 2,19E **Elevation:** 120 masl **Sampling time:**. Jan 1926 – Nov 1927 **Samples**: 27 (one selected: 4th Dec 1926, strong wind) **Meteorolog. Parameters: :** - temperature,wind, weather **Station 612**	**Pettenkofer variant after Vandenberghe** Passing air though baryta water and titration **Remarks:** Only one value selected : altitude 120 m, strong winds **Error:** +-1-2 %
	Investigator: August Krogh, Prof., zoology university of Kopenhagen, Nobel awardist 1920; Dr. Odum **Location:** Greenland /Danmark **Elevation:** - **Sampling time:**. summer 1926 **Samples**: - **Meteorolog. Parameters: :** - Station 613	**Pettenkofer variant** Passing air though baryta water and titration by HCl **Remarks:** **Error:** +-3 %
1927	**Investigator: Robert Gut**, Dr., forest engineer Zurich **Location:** Zuerichberg, forests near Zurich, 4 stations, **Elevation:** 622, 619, 642, 646 masl, 34, 28, 23, 18, 13, 10, 5, 1 m altitude+ more altitudes **Sampling time:**. July 1927 – July 1928 **Samples**: 5000 **Meteorolog. Parameters: :** - temperature,wind, weather Station 617	**Volumetric apparatus by Gut** similar to Haldane apparatus absorption of air in KOH, manometric reading of changed air volume over solution temperature and pressure controlled as Krogh apparatus **Remarks:** Temperature sensible, too low levels at higher temperature errors in CO2 absorption heat, large diurnal variation the same as measured in modern with somewhat higher CO2 levels; no calibration against Pettenkofer **Error:** +-2-3%

	Investigator: Donald D. van Slyke, Prof. Dr., biochemist **Location:** Rockefeller Institute laboratory , New York **Elevation:** **Sampling time:**. 1927 **Samples**: >2 **Meteorolog. Parameters: : -** temperature Station 618	**Volumetric apparatus by Van Slyke (manometric)** Absorption of air in NAOH Manometric reading of pressure change **Remarks:** Comparison with the Haldane apparatus **Error:** +-1 %
1930	**Investigator: Paul Lehmann**, Dr., Klimatologie der Hochschule für Bodenkultur **Location:** Lunz, biological station Seehof lat 47,8N, 15E **Elevation:** 1280 masl + 2m **Sampling time:**. 1927 **Samples**: >44 soil and air (29) **Meteorolog. Parameters: : -** temperature , pressure, wind, weather Station 619	**Pettenkofer variant Krogh/ Rehberg 1928** Passing air though baryta water and titration by HCl **Remarks:** Measurements over crops, 2, over Topinambur; photosynthesis absorption reduction: **Error:** +-3 %
1931	**Investigator: Donald D. van Slyke**, Prof. Dr., biochemist **Location:** Rockefeller Institute laboratory , New York lat 40,30N, -45,10E **Elevation:** 8 masl **Sampling time:**. 1931 **Samples**: >2 **Meteorolog. Parameters: : -** temperature Station 620	**Volumetric apparatus by Van Slyke (manometric)** Absorption of air in NAOH Manometric reading of pressure change **Remarks:** Comparison with the Haldane apparatus **Error:** +-1 %
1932	**Investigator: Kurt Buch**, Prof. Dr., chemistry Institute of ocean research, Helsinfors **Location:** northern Atlantic ocean lat 56,41N 5,5E, 55,56N, -6,21E, 59,07N, -6,14E, 62,01N, -5,3E, 64,5N, -10,4E, + locations at the northern icelandic coast **Elevation:** 8 masl **Sampling time:**. summer 1932 (7th July – 9th Sept) **Samples**: 28 (double determinations) **Meteorolog. Parameters: : -** temperature, weather Station 632	**Pettenkofer variant Krogh/ Rehberg 1928** Variant by Buch Passing air though baryta water and titration by HCl **Remarks:** Sampling over sea surface: **Error:** +-2 %
1933	**Investigator: Kurt Buch**, Prof. Dr., chemistry Institute of ocean research, Helsinfors **Location:** Liinahamari, Petsamofjord (now Russian) lat 69° 38' 29 N, Longitude: 31° 20' 36 E **Elevation:** 8 masl **Sampling time:**. 31 October 1933 – 4 May 1935 **Samples**: weekly, 52 (double determinations) **Meteorolog. Parameters: : -** temperature, wind, weather Station 633 **Investigator: Thomas Moyer**, Dr., chemistry AMERICAN SMELTING AND REFINING COMPA.NY, Utah , USA **Location:** 5 km out of Salt Lake City, Utah, lat 69°	**Pettenkofer variant Krogh/ Rehberg 1928** Variant by Buch Passing air though baryta water and titration by HCl **Remarks:** Sampling over sea surface: **Error:** +-2 % **Thomas Moyer Autometer** Absorption of CO2 from air stream with 300 ccm/min in 0,005N NaOH**;** measuring the electrical conductance of the solution at constant temperature

	40 N, Longitude: -111 E **Elevation:** ~1900 masl **Sampling time:**. 19 Jan 1933 – 13 Feb 1933 **Samples**: >135 (continous air stream) **Meteorolog. Parameters:** - Station 634	with a recording Wheatstone bridge, making use of the fact that sodium hydroxide solution has about twice the conductance of the equivalent sodium carbonate solution. Temperature thermostat controlled Source: Air pollution and plant life J. N. B. Bell,Michael Treshow, p.7; Wiley 2002 **Remarks:** One of the first automatic systems, calibration against Pettenkofer titrimetric method: difference max+-0,3-0,6% **Error:** +-1 %
1934	**Investigator: John.G. Waugh**, Dr., physicist Cornell university Ithaca , USA **Location:** Ithaca, Plant science building, roof, rural area **Elevation:** 135 masl **Sampling time:**. 11 April 1934 – 11 Oct 1934 **Samples**: >37 **Meteorolog. Parameters:** Station 635	**Thomas Moyer Autometer variant** Absorption of CO2 from air stream with 350 ccm/min in 0,00488N NaOH**;** measuring the electrical resistance of the solution at constant temperature with a recording Wheatstone bridge. 2,5 l air samples in flasks, 2-3 determinations/hour **Remarks:** Double determination in outside air **Error:** average +-1 %
1935	**Investigator: Yrjö Kauko**, Prof. Dr., chemist University of Helsinki **Location:** over Helsinki, by aeroplane, vertical profile **Elevation:** 0-1500 masl **Sampling time:**. 20 Feb 1935, 7 Dec 1935, 13 Dec 1935 **Samples**: 15 **Meteorolog. Parameters:** Temperature, wind, weather Station 636 **Investigator: J.B.S. Haldane**, Prof. Dr., chemist University of Helsinki **Location:** Scotland rural and coast, **Elevation:** 1,2-21 masl **Sampling time:**. July - December 1935 **August:** Cloan (Perthshire**) day/night** **Samples**: 1500 153 samples at Cloan (Perthshire)= 324 ppm day/386ppm night; samples coast of Ayreshire (Aug/Sept)= 370 ppm wind from the sea; night air at Oxford in October to Nov= 330 ppm av and 380 av; **Meteorolog. Parameters:** wind, weather Station 639	**Kryogenic Condesation Method** 30 l air was passed through conc. H_2SO_4., stream of 10l/std measured by difference manometer, passed though cooling coils, and sampled in flasks filled with $CaCl_2$ and determined by weighting the volume of the displaced solution. **Remarks:** Most accurate method Calibration against potentiometric method (+-1%) and Pettenkofer **Error:** average +-0,33 % **Haldane volumetric**, improved See above **Remarks:** All pipettes in the same water bath Also measurements of CO2 from soils and combustion (London) **Estimated error:** modified +--2,5 %
1936	**Investigator: Kurt Buch**, Prof. Dr., chemistry Institute of ocean research, Helsinfors **Location:** travel to Spitsbergen lat 69° N - 80N, lon 0°E- 20 E **Elevation:** 8 masl **Sampling time:**. 14 Aug – 24 Aug 1936 **Samples**:, 13 in flasks **Meteorolog. Parameters: :** - temperature, wind, weather Station 652	**Pettenkofer variant Krogh/ Rehberg 1928** Variant by Buch Passing air though baryta water and titration by HCl **Remarks:** Sampling over sea surface: **Error:** +-2 %

	Investigator: Johann Ulrich Duerst, Prof. Dr., veterinary science Institute veterinary hygiene University of Bern (Switzerland) **Location:** Ins (near Bern Switzerland**)** lat 47° N, lon 7,1 E , rural area **Elevation:** 499 masl **Sampling time:**. 1936-1938, max. 6 samples/day **Samples**:, 1000 **Meteorolog. Parameters: :** temperature, wind, weather Station 653	**Pettenkofer variant Hesse** Calibration with Pettesson Sonden analyser Passing air though baryta water and titration by HCl **Remarks:** Sampling 1m over meadow surrounded by:shrubs and trees **Error:** +-3 %
1938	**Investigator: Kurt Buch**, Prof. Dr., chemistry Institute of ocean research, Helsinfors **Location:** Baltic sea, lat 65 -50, lon 17-29 ; see separate file **Elevation:** 0 masl **Sampling time:**. 1927-1938, day night **Samples**:, 185 at 185 different stations **Meteorolog. Parameters: :** temperature, oceanographic parameters Station 838	**Pettenkofer variant Krogh/ Rehberg 1928** Variant by Buch Passing air though baryta water and titration by HCl **Remarks:** Sampling over sea surface: **Error:** +-2 %
1939	**Investigator: Wilhelm Kreutz**, Dr., chemistry Director of the weather station at Giessen (Germany), Reichswetterdienst **Location:** Giessen periphery, near rural area **Elevation:** 499 masl **Sampling time:**. August 1939- January 1941, 16 samples per day, otherwise samples at 7:00, 14:00, 21:00 at 4 altitudes; (total 550 days) **Samples**:, >30 000; > 25 000 used soil samples: 1647, special CO2/temperature analyses: 1098 + analysis in higher air layers + 2176 diurnal sampling every 1,5 hours+ preexaminations at Heidelberg; 7054 in 1939, 16773 in 1940, 1429 in 1941 **Meteorolog. Parameters: :** temperature, wind, precipitation, radiation, pressure, humidity, cloud coverage, weather Station 839 **Investigator: J. Verduin,** Dr., botany; IOWA state university **Location:** IOWA Agricultural experimental station; cornfield, lat 42,024N, -93,6E **Elevation:** 299 masl, 1 m **Sampling time:**. July/Aug 1939/40 **Samples**:1939: 53; 1940: 203 **Meteorolog. Parameters:** - Station 840	**Manometric Riedel C gas analyser designed by Schuftan** 0,5 l flask sampling , Pressing air sample through KOH by turning vessel by 90 degree, reading volume change by capillary manometer **Remarks:** Sampling in 4 altitudes: 0, 0,5, 2, 14 m; monitoring radiation, precipitation, cloud cover, wind speed, pressure, humidity. Temperature, CO2 Analysis in temperature controlled room **Error CO2:** +-1,5 % **Pettenkofer** according to Heinicke and Hoffmann, modified Absorption of CO2 in NaOH, precipitation in BaCl2, titration with HCl, phenolphthalein **Remarks:** 25% absorption at 1 m over cornfield from normal level **Error:** 3%
1940	**Investigator: Wilhelm Bazett**, Prof. Dr., physiology; University of Pennsylvania, Philadelphia **Location:** Philadelphia **Elevation:** 105 masl **Sampling time:**. 1940 **Samples**: 9 **Meteorolog. Parameters:** - Station 841	**Haldane volumetric gas analyser variant** See above **Remarks:** No date and other parameters available **Estimated error:** modified +--13 %
1941	**Investigator: Ernest Earl Lockhart,** Dr.,	**Haldane volumetric portable gas analyser**

	physiologist, Harvard university, Boston USA, **Arnold Court**, meteorologist, US weather Bureau **Location:** Antarctica, West Base 78,29S, -163,5 E **Elevation:** 244 masl **Sampling time:**. July 1940 –Jan 1941 **Samples:** 26 **Meteorolog. Parameters:** wind, temperature, pressure, weather, O2 Station 842 **Investigator: Harry J. Fuller,** Prof., botanist, University of Illinois **Location:** Campaign county forest and grassland**,** Illinois, Urbana 40,1N, -88,1 E **Elevation:** about 214 masl **Sampling time:**. 22 June 1941 –9 July 1941, 1pm **Samples:** 144 (4 locations) **Meteorolog. Parameters:** - Station 846	See above **Remarks:** Calibrated according to van Slyke **Estimated error:** +-5% **Haldane volumetric gas analyser** 100ml glass tubes, analysed in laboratory **Remarks:** Quadrouple determinations **Estimated error:** +-2,5%
1943	**Investigator: R.K. Misra,** Dr., chemist, Central Agricultural Meteorological Observatory Poona , India **Location:** experimental fields near Poona, rural area; lat 18,5N, 73,8 E **Elevation:** about 555 masl **Sampling time:**. Dec 1941 –Nov 1943, **Samples:** atmosphere >1532; soil > 1500 **Meteorolog. Parameters:** temperature, wind, precipitation Station 847	**Pettenkofer** Passing air though baryta water and titration by oxalic acid, Phenolphthalein as indicator **Remarks:** Flask samples anylsed in laboratory Double determinations, difference in the 3^{rd} decimal **Error:** +-4 %
1944	**Investigator: Eugen Glückauf,** Dr., chemist, University Science Laboratories Durham (UK) **Location:** 4-10km over England; Kew garden; lat 51,5N; 0,29W **Elevation:** about 4000-1000 masl; Kew Garden 15 masl **Sampling time:**. winter/spring 1944, **Samples:** 12 in troposphere, several near ground London **Meteorolog. Parameters:** - Station 849	**Condensation method** Freezing out CO2 and H20 by liquid air, measuring the pressure of CO2 **Remarks:** Author states CO2 in water was negligible, no check **Error:** 10% (see reference p. 621; 0,001% CO2)
1946	**Investigator: Per F. Scholander, Prof.** Dr., botanist, 1946 at Edward Martin Biological Laboratory, Swarthmore College Swarthmore **Location:** Swarthmore lat 39.90N, -75.35E , rural area **Elevation:** Edward Martin laboratory 77 masl **Sampling time:**. 1946 **Samples:** >3000 **Meteorolog. Parameters:** - Station 850	**Manometric Scholander Analyser** Absorption of air in KOH, automatically measuring pressure change **Remarks:** checked against Haldane analyser: same accuracy **Error:** paper 1946 +- vol 0,015% stddev of tests 58 (mean 349)=16,5% improved analyser=
1947	**Investigator: Per F. Scholander, Prof.** Dr., botanist, 1947 at **Location:** Point Barrow Arctic Laboratory lat 71,3N, -156,64W, **Elevation:** 2 masl **Sampling time**: Oct 1947 – March 1949 **Samples:** :. ~350 **Meteorolog. Parameters:** O2 Station 851	**Manometric Scholander Analyser** Absorption of air in KOH, automatically measuring pressure change **Remarks:** checked against Haldane analyser: same accuracy **Error:** paper 1946 +- vol 0,015% stddev of tests 58 (mean 349)=16,5% improved analyser=

1949	**Investigator: H. W. Chapman,**Dr., botanist, University of Nebrasca, Lincoln **Location:** near Alliance Nebrasca, rural area lat 42,4N, -103E **Elevation:** 1220 masl **Sampling time**: June-Aug 1949 -1951 **Samples: :**. 1949: 220 ; 1950: 148, 1951: 177 **Meteorolog. Parameters:** temperature Station 852	**Pettenkofer** according to Heinicke and Hoffmann, modified; Absorption of CO2 in KOH, precipitation in BaCl2, titration with HCl, phenolphthalein **Remarks:** Control tower 10-30 cm over potato plants; 28% variability in CO2 absorption by potato leaves Absorption by potato plants: average 14%, **Error:** 3%
1950	**Investigator: Hal R. de Selm,**Prof., botanist, Ohio state university; later University of Tennessee, Knoxville **Location:** near Blacklick Creek Ohio, rural area lat 40,1N, -82,4E **Elevation:** 327 masl, 0,9m, 9,m, 12m above ground **Sampling time**: April - July 1950 , 4 hourly at day **Samples: :**.>127 **Meteorolog. Parameters:** temperature, pressure, humidity, wind Station 853	**Pettenkofer** according to Heinicke and Hoffmann, modified by Böhning 1948; http://www.plantphysiol.org/cgi/reprint/24/2/222 Absorption of CO2 in KOH, precipitation in BaCl2, titration with HCl, phenolphthalein **Remarks:** Measurements in forest **Error:** 3%
	Investigator: Karl Egle,Prof., botanist, Director of the institute of applied botany Hamburg, later on president of the botanical society of Germany **Location:** Frankfurt botanical institute lat 50,1N, 8,6E **Elevation:** 348 masl, + 6-10 m **Sampling time**: 19 may 1950, 5:00AM-1:30PM **Samples: :**.>1620, **Meteorolog. Parameters:** temperature, pressure, humidity, wind; rainy day Station 853	**URAS** continous sampling and plotting (NDIR),drying air by streaming through silica gel ; infrared absorption by CO2 **Remarks:** Continuous gas stream; sample analysis within every 30 sec **Error:** < +- 1% (0,0001 Vol%)
	Investigator: Siegfried Strugger, Prof., botanist, University Muenster (Germany) **Location:** botanical garden university Muenster, rural area, grassland lat 51,9N, 7,61E **Elevation:** 65 masl; 3-4m above ground **Sampling time**: 18/29 July-1950, 8:00am-6am **Samples: :**. > 2640 **Meteorolog. Parameters:** weather Station 854	**URAS** continous sampling and plotting (NDIR),drying air by streaming through silica gel ; infrared absorption by CO2 **Remarks:** Continuous gas stream; sample analysis within every 30 sec **Error:** < +- 1% (0,0001 Vol%)
1951	**Investigator: Bruno Huber,** Prof., botanist, forest and botanical institute, University Munic (Germany) **Location:** over wheat field in Duernast (48,4N, 11,691E) Freising agricultural faculty Weihenstephan TU Munic **Elevation:** 476 masl; 50cm-1,2m, 4,5m, 22,5 m beside the field, 50-100m balloon **Sampling time**: 15 May- 14 Aug 1951 **Samples: :**. > 15 000 **Meteorolog. Parameters:** weather, temperature, rain Station 855	**URAS** continous sampling and plotting (NDIR),drying air by streaming through silica gel ; infrared absorption by CO2 **Remarks:** Continuous gas stream; sample analysis within every 30 sec **Error:** < +- 1% (0,0001 Vol%)
1952	**Investigator: H. W. Chapman,** Dr., botanist,	**Pettenkofer** according to Heinicke and Hoffmann,

	Iowa state College, Ames, Iowa **Location:** cornfield, Iowa state college television tower, rural area lat 42,4N, -103E **Elevation:** 1220 masl; 1m, 20m above corn field ; TV tower 10m, 30m, 152m **Sampling time**: July-Oct1952, 3AM-midnight **Samples: :.** > 66 **Meteorolog. Parameters:** temperature, pressure wind Station 856	modified; Absorption of CO2 in KOH, precipitation in BaCl2, titration with HCl, phenolphthalein **Remarks:** **Error:** 3%
1954	**Investigator: Stig Fonselius,** Dr., meteorologist, Institute of marine research Helsingfors Finland **Location:** 15 stations throughout Scandinavia; data from Plönninge Sweden lat 56,8N, 12,9E and Tvärminne Finland 59,8N, 22,9E selected **Elevation:** Plönninge 59; Tvärminne 14,9 masl **Sampling time**: Nov 1954-Dec 1959, 1st,10th, 20th of each month , 1PM **Samples:** > 2700 **Meteorolog. Parameters:** temperature, wind, pressure, weather Station 851	**Pettenkofer variant by Krogh/ Rehberg** 1929 Passing a small amout of air through Ba(OH)2; titration by HCl see above **Remarks:** air samples collected in flasks analysed in laboratories at Stockholm and Sweden 6 sampling location2 in Sweden, 4 in Finland, 3 in denmark, 2 in Norway; only data from Plönninge Sweden and Tvärminne (Finland seashore) used **Error:** +-3 %
1955	**Investigator: Charles Keeling,** Dr., chemist, California institute of Technology , Pasadena USA **Location:** see below **Elevation:** 1. 10 masl +1 m; 2. 1950 masl 1m 3. 2500 masl; 4. 3000 masl; 5. 3800 masl; 6. 4000 masl 7. 4300 masl; 8. 1900 masl 9. 0 masl; 10. 170 masl **Sampling time**: 1.Big Sur State Park: 18-19 May 55 2. Yosemite National Park, Tamarack: 2-3 June 1955 3. Yosemite National Park, Lake Tenaya: 3 June 1955 4. Yosemite National ParkTioga Pass: 3 June 1955 5. Inyo Mountains,: Mt. Barcroft 8 July 1955 6. Inyo Mountains, Mountain devide: 8-9 July 1955 7. Inyo Mountains, white Mountain summit: 9 July 1955 8. Hart´s Pass 1 Sept 1955 9. Olympic National Parc Beach trail: 6-7 Sept 1955 10. Olympic National Hoh River Ranger st.: 6-7 Sept 1955 11. Rock Lake 10-11 Sept 1955 + further locations see reference in 1956 **Samples:** 50 **Meteorolog. Parameters:** temperature, wind,	**Cryogenic condensation method** Condensation of sample by liquid nitrogen, freeing the condensate from water by fractionated distillation and identifying gas by mass spectroscopy and determination quantity by a manometer. **Remarks:** air samples collected in 5 ltr. flasks analysed in laboratory, additionally determination of C13 isotope **Error:** +-1 %

	pressure, humidity, weather Station 862	
1956	**Investigator: Charles Keeling,** Dr., chemist, Scripps Institute of Oceanography, USA **Location:** see below 1. Coastal Redwood Canyon, 6-7 June; 150 masl 2. Coastal Redwood Valley 5-6 June; 70 masl 3. Sierra Nevada Pine and Fir Forest California 10-11 June 1956; 1950 masl 4. Mogollan Rim Pine Forest Arizona; 16-17 May 1956; 2100 masl 5. Borrego desert; 02. Feb 56, 340masl 6. Inyo Mountains; 9-14 March 1956; 3800 masl 7. Sonora desert; 21-22 April 1956, 550 masl **Elevation**: 1-7: 70 -3800 masl see above **Sampling time**: See above **Samples:** 51 **Meteorolog. Parameters:** - Station 869	**Cryogenic condensation method** Condensation of sample by liquid nitrogen, freeing the condensate from water by fractionated distillation and identifying gas by mass spectroscopy and determination quantity by a manometer. **Remarks:** air samples collected in 5 ltr. flasks analysed in laboratory, additionally determination of C13 isotope **Error:** +-1 %
1957	**Investigator: Ferdinand Steinhauser,** Prof., meteorologist, Zentralanstalt für Meteorologie und Geodynamik, Vienna, Austria **Location:** weather station Hohe Warte Vienna, Austria, lat 48,25N, 6.35E **Elevation:** 205 masl +25 m tower **Sampling time**: Jan 1957- June 1958, daily 1PM **Samples:** 546 **Meteorolog. Parameters:** wind, (temperature, pressure, weather: other data set) Station 870	**Pettenkofer variant by Krogh/ Rehberg** 1929 Passing a small amout of air through Ba(OH)2; titration by HCl see above **Remarks:** Air collected from the tower of the institute and analysed in laboratory **Error:** +-2 %
1959	**Investigator: Walter Bischof,** Dr., engineer University of Stockholm, **Location:** near/ over Stockholm, Sweden lat 48,25N, 6.35E; Dalarö: lat 59.13N, 18.4E (island) Bryggan:, lat 59.78N, 14.13E, Golf course 1 km away, Djusrholm **Elevation:** 100 m -3 km masl over Stockholm; Dalarö: 0 masl; Djursholm: 16 masl, Bryggan: 154 masl **Sampling time**: 28 October 1959 over Stockholm, 11-13 Aug and 15-17 Sept 1959 at Golfbanan, 15/16 Aug Dalarö , 28 July at Bryggan and a golf course 1km away (1 Sept, 8 Dec.) **Samples:** 92 **Meteorolog. Parameters:** eastern winds Station 875	**NDIR gas analyser** **Remarks:** used since May 1959 **Error:** +-1-4 %

Note: Montsouris 1877-1910