

## Supplementary Material

### CSIRO High-precision Measurement of Atmospheric CO<sub>2</sub> Concentration in Australia.

#### Part 1: Initial Motivation, Techniques and Aircraft Sampling

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#### Introduction

**Table S1.** Summary of key international CO<sub>2</sub> measurement programmes. This is not a complete list of contributors but summarizes the earlier and more sustained programmes. For more detail and for the global flask sampling network, see Pearman (1980), the Reports of the Global Atmospheric Watch (e.g. WMO, 1991, 1993) and the international CO<sub>2</sub> data archives (Carbon dioxide Information Analysis Center; <http://mercury.ornl.gov/cdiacnew/>; World Data Centre for Greenhouse Gases; <http://ds.data.jma.go.jp/gmd/wdcgg/>).

Nation	Location	Start date	Selected references	
<b>Australia</b>	Cape Grim	145°E 41°S	1976	Beardsmore and others (1984)
	Macquarie Is.	159°E 54°S	1979	Beardsmore and others (1984)
	Mawson	61°E 68°S	1977	Beardsmore, Pearman, O'Brien. (1984)
	South-west Pacific		1972	Garratt and Pearman (1973); Pearman and Garratt (1973); Pearman and Beardsmore (1977); Beardsmore and others (1978)
<b>Canada</b>	Alert	62°W 82°N	1975	Wong (1984)
	Sable Is.	60°W 44°N	1975	Wong (1984)
	Weather ship P	145°W 50°N	1969	Wong (1984)
<b>France</b>	Amsterdam Is.	78°E 38°S	1980	Gaudry, Ascensio, Lambert. (1983); Ascensio-Parvy, Gaudry, Lambert (1984)
<b>Germany</b>	Schauinsland	8°E 48°N	1967	Levin, Graul and Trivett (1995)
<b>Italy</b>	Mt. Cimone	11°E 44°N	1978	Ciattaglia (1983); Ciattaglia, Cundari, Colombo (1987)
<b>Japan</b>	Syowa	40°E 69°S	1984	Tanaka, Nakazawa, Aoki (1983); Tanaka and others (1987)
	North-west Pacific		1978	Nakazawa and others (1991)
<b>New Zealand</b>	Baring Head	175°E 41°S	1972	Lowe (1974); Lowe, Guenther, Keeling (1979)
<b>Sweden</b>	North Atlantic		1961	Bischof (1865, 1971, 1973, 1975)
<b>USA</b>	Mauna Loa	156°W 20°N	1960	Pales and Keeling (1965); Keeling and others. (1976a); Peterson and others (1977)
	Barrow, Alaska	157°W 71°E	1961	Kelly (1969); Peterson and others (1982)
	South Pole	90°S	1957	Bronn and Keeling (1965); Keeling and others (1976b)
	Samoa		1973	Komhyr and others (1985)

## CO<sub>2</sub> Concentration Measurements over Crops and Forests in Australia—Post 1970

### *Main Results (Aerodynamic Method)*

The vertical transfer of CO<sub>2</sub> above a crop occurs primarily through the action of turbulence, with the vertical flux given by (e.g. Webb *et al.* 1980 – their Eqs. 15 and 16)

$$F_c = \overline{w\rho_c} \quad (1)$$

where  $w$  and  $\rho_c$  are the instantaneous vertical velocity and CO<sub>2</sub> density respectively. Here the overbar represents a time average (usually 0.5 to 1 h). This is the true surface flux, and to a high degree of approximation can be written as

$$F_c/\rho_a = \overline{w'c'} \quad (2)$$

where  $\rho_a$  is the mean density of dry air, the variable  $c$  is the CO<sub>2</sub> concentration or mass mixing ratio (see below) and the primed quantities are fluctuations from their mean

In the early 1970s the eddy covariance in Eq.2 could not be measured and so a flux-gradient relationship valid in the surface layer was used. In modern terminology this can be written using the Monin-Obukhov similarity theory (e.g. see Garratt 1994, Chapter 3). For steady-state and horizontally homogeneous conditions the vertical flux,  $F_s$ , of a property  $s$  is related to its vertical gradient via

$$F_s/\rho = -K_s \partial s / \partial z \quad (3)$$

where  $\rho$  is the mean air density and  $K_s$  is an eddy diffusivity given by

$$K_s = k u_* z / \varphi_s(z/L) \quad (4)$$

Here the von Karman constant,  $k = 0.4$ ,  $u_*$  is the friction velocity,  $z$  is height,  $L$  is the Obukhov length, and the  $\varphi(z/L)$  functions represent the effect of thermal stratification on the vertical transfer of any property  $s$ . Their empirical forms are now well-known, in part due to the work of Swinbank, Dyer, Webb and Hicks at CSIRO's Division of Meteorological Physics in the 1950s and 1960s (e.g. Swinbank and Dyer 1967; Dyer and Hicks 1970; Webb 1970). Both  $u_*$  and  $\varphi(z/L)$  can be evaluated from wind speed and temperature profiles. In fact, for the Rutherglen data we use Eq. 3 in finite difference form since wind speed and temperature were available as vertical differences between heights of 1 m and 2 m above the crop (wind speed in fact was measured at several heights but not air temperature). For wind speed ( $u$ ),  $F_s/\rho = u_*^2$  so that Eq.3 can be written as,

$$(kz/u_*) \partial u / \partial z = \varphi_m(z/L) \quad (5)$$

and for the CO<sub>2</sub> flux (cf. Eq. 2),

$$F_c/\rho_a = -K_c \partial c / \partial z \quad (6)$$

with  $K_c$  given by Eq. 4 in terms of  $\varphi_c(z/L)$ . Thus we finally need to evaluate  $u_*$  and  $z/L$  and to do this we introduce the Richardson number,  $Ri$ ,

$$Ri = 0.033[\partial\theta/\partial z][\partial u/\partial z]^{-2} \quad (7)$$

where the numerical factor 0.033 is simply  $g/\theta$ ,  $g$  being the acceleration due to gravity and  $\theta$  is mean potential temperature. It is well-known that, for  $\partial\theta/\partial z < 0$  ( $Ri < 0, z/L < 0$ ),  $z/L = Ri$  and for  $\partial\theta/\partial z > 0$  ( $Ri > 0, z/L > 0$ ),  $z/L = Ri/(1 - 5Ri)$ , and that

$$\varphi_c(z/L) = [[\varphi_m(z/L)^2] = (1 - 16Ri)^{-0.5} \quad (8)$$

for  $Ri < 0$ , and

$$\varphi_c(z/L) = \varphi_m(z/L) = [1 - 5Ri]^{-1} \quad (9)$$

for  $Ri > 0$ .

Taking the differential form for a vertical height increment  $\Delta z$ , we finally have for the vertical flux of  $\text{CO}_2$ ,

$$F_c/\rho_a = -m\Delta u\Delta c \quad (10a)$$

and

$$m = [kz/\Delta z]^2 / [\varphi_c \varphi_m], \quad (10b)$$

where  $z$  is the geometric mean between heights 1 and 2.

For the Rutherglen data we have  $\Delta z = 1$  m (for levels 1 m and 2 m above the crop), and  $z_2 = 2 + h - d$ ,  $z_1 = 1 + h - d$ ,  $h$  being the crop and  $d$  the zero-plane displacement, taken as  $d = 2h/3$ . For  $h = 0$ ,  $z = 1.41$  m; for  $h = 0.3$  m,  $z = 1.52$  m,  $d = 0.2$  m,  $z = 1.625$  m; for  $h = 0.9$  m,  $d = 0.6$  m,  $z = 1.73$  m.

Note that  $u_*$  is evaluated here using the finite difference form of Eq.3, using  $\Delta u$  from the wind-speed measurements, as compared with using the integrated form of Eq. 3, using  $u$  at height  $z$  and an assumed value for the aerodynamic roughness length  $z_0$ . We have chosen the differential form so that this extra assumption is not required ( $z_0$ , as with  $d$ , is a function of  $h$  and hence of time).

To assess the reliability of this approach (using  $\Delta u$ ) we took the integral form of Eq. 3 for neutrally stratified conditions

$$ku/u_* = \ln z/z_0 \quad (11)$$

so that  $F_s$  can be calculated from

$$F_c/\rho_a = -m'u\Delta c \quad (12a)$$

with

$$m' = [k^2 z/\Delta z]/\ln z/z_0 \quad (12b)$$

Actually we took six individual times from the days shown in Fig.1 for which  $-0.005 < Ri < 0.005$  and evaluated  $F_c$  using wind speed  $u$  at a single level. Thus, values of  $z_0$  that gave identical values of  $F_c$  from both differential and integral approaches were found to be 0.01 m (24 July), 0.03 m (7 August), 0.06 and 0.07 m (26 September) and 0.06 and 0.07 m (14 October). The

corresponding values of  $z_0/h$  varied between 0.05 to 0.1, typical of crops and lending confidence to the  $F_c$  values evaluated throughout the season.

## **CSIRO Aspendale Aircraft Sampling Programme: 1972–92**

### ***Sampling Techniques***

In late 1971 initial discussions were undertaken with the Department of Civil Aviation with a view to sampling air from their weekly training flights in a Fokker Friendship (F-27). The collection of air samples commenced in March 1972 over Bass Strait and south-eastern Australia using manually operated equipment carried on the F-27. Subsequently, routine flights at weekly or bi-weekly intervals were made to Tasmanian and several mainland airports, at altitudes up to 5 km. When the F-27 was unavailable, or when synoptic conditions were of special interest, flights were supplemented using several types of chartered light aircraft up to 1992.

In early 1972, after earlier discussions with Qantas Airways, which led to further development of sampling equipment for automatic operation by the on-board navigator, the aircrew began sampling over the Tasman Sea, at the cruise levels of Boeing 707 aircraft flying on routine commercial flights between Melbourne and New Zealand. On these flights, at intervals of two to three weeks, samples were collected between heights of 9 and 13 km, and thus occasionally in the lower stratosphere. CSIRO personnel also collected samples, at intervals of 8 to 10 weeks, from Boeing 747 aircraft operating between Sydney and Melbourne on training flights. On most of such flights samples were collected at various altitudes as aircraft climbed to, or descended from, their operating altitude of 11–12 km.

Discussions with Trans Australia Airlines in 1972 and early 1973 led to their agreeing to the periodic installation of a 5 L air sampling system mounted into the tail engine of a Boeing 727 aircraft, automatically sampling at cruise altitude (between 8.5 and 13 km) on commercial flights between Melbourne and Sydney, Brisbane and Perth. Regular sampling over the Great Australian Bight commenced in December 1973, continuing throughout the 1970s and much of the 1980s over and around the Australian continent, including the Tasman Sea. The sampling programme was replaced in 1989 by an international coordinated programme (see main text).

On all aircraft, with the exception of the Boeing 727, air was pumped from a cabin air conditioning outlet and collected in glass flasks of nominal volume 0.5 L. Sets of 6 to 10 flasks were fitted into special carrying cases for safe transportation and ease of access to individual flasks inside aircraft. Air was dried by passing it through a tower of magnesium perchlorate prior to entering each flask, and then each flask was filled to an overpressure of one atmosphere (100 kPa), but only after 2 to 3 min of pumping to ensure a complete purging of the system. After each flask was filled, time, aircraft location and altitude were noted.

On the Boeing 727 aircraft sampling involved a self-contained automatic unit fitted into the tail section. This unit connected directly into the main air-conditioning duct, with air drawn into a cadmium plated steel cylinder of volume 3.8 L. After aircraft ascent through a pre-set altitude (typically 8.5 km) an altitude-sensitive (pressure) sampling valve activated a time delay, the

delay being determined prior to each flight to allow the aircraft to reach cruise altitude and enter the area from which the air sample is required. After the delay time had expired inlet and outlet valves were opened, the cylinder flushed for 3 min at about 12 L min<sup>-1</sup>, the outlet valve then closed and a pressurised sample taken. Data were later provided by the Department of Transport to allow the time, location and height of each sample to be calculated.

### ***Tropospheric Profiles and Bulk Eddy Viscosity (K)***

We make use of the mean Northern Hemisphere vertical profiles in Stephens *et al.* (2007, Figs.1 and 3) and deduce an effective bulk tropospheric  $K$  value by using published carbon fluxes, essentially available north of about 30°N. This allows us to assess the validity of earlier assumed  $K$  values and that assumed in Garratt and Pearman (1973a).

- (i) For winter, the estimated gradient of  $-0.35$  ppm km<sup>-1</sup> (viz. a mean decrease of 2.3 ppm from heights of 1.5 and 8 km) implies a net surface source. Now Fig.7 of Pearman *et al.* (1983) and Fig.1 of Miyazaki *et al.* (2008) suggest a net winter flux of about  $2 \times 10^{-9}$  kgC m<sup>-2</sup> s<sup>-1</sup> (for 1980 – the former and for 2000 – the latter), and combined with the above concentration gradient gives  $K = 20$  m<sup>2</sup> s<sup>-1</sup>, noting that we have converted from kgCO<sub>2</sub> to kgC.
- (ii) For the mean annual profile, the estimated vertical gradient of  $-0.07$  ppm km<sup>-1</sup>, again implies a net surface source. Using Fig.7 of Pearman *et al.* (1983) and now Fig.8 of Le Quere *et al.* (2016) suggests a net annual flux of about  $1 \times 10^{-9}$  kgC m<sup>-2</sup> s<sup>-1</sup> (for years 1980 and 1990–95 respectively), and combined implies  $K = 50$  m<sup>2</sup> s<sup>-1</sup>, somewhat larger than earlier estimates and assumptions.
- (iii) Finally we use the winter and annual concentration gradients given in Fig.3 of Stephens *et al.*, valid for the height range 1 to 4 km, and with the above fluxes yields a  $K$  range of 10 to 20 m<sup>2</sup> s<sup>-1</sup>. These lower estimates of  $K$  merely reflect the use of a shallower layer depth (1 to 4 km heights) and a lower mean height, consistent with Bolin and Bischof (1970).

Taking the above estimates into account, together with the range suggested by Bolin and Bischof (1970), we are inclined to use  $K = 25 (\pm 10)$  m<sup>2</sup> s<sup>-1</sup> for the Southern Hemisphere troposphere and for seasonally-averaged profiles.

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