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Summary The multiple tracer gas technique of I'Anson *et al.*¹ has been improved, in order to increase the rate at which samples can be taken. Using parallel gas chromatographic separation columns, in conjunction with an electron capture detector, it is now possible to take an air/tracer gas sample every 30 seconds in the case of a 2-zone ventilation and air movement test. Rapid sampling enables a new, simplified analysis of the air movement between two connected zones to be employed. This analysis derives ventilation rates and intercell airflows simultaneously. A specimen set of results for 2 cell ventilation/air movement is given.

The measurement of airflows using a rapid response tracer gas technique

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List of symbols

N_x	ventilation rate in cell x	air changes per hour
Q_x	amount of air flowing from cell x to outside	m^3/h
F_{xy}	air flow from cell x to cell y	m^3/h
C_{Ax}	concentration of tracer gas A in cell x	ppm
C_{OAx}	concentration of tracer gas A in cell x at time = 0	ppm
t	time	h

1 Introduction

1.1 Single cell model

The measurement of ventilation rates and internal airflows by tracer gas techniques, as originally proposed by Dick² has been developed and extended by several workers, for example Sinden³, Sherman⁴, Perera⁵ and I'Anson *et al.*¹ for the determination of multi-cell air movements. This paper is concerned with the development and refinement of the work of I'Anson *et al.* in terms of data collection and analysis.

The analysis of single volume tracer gas decay (assuming good mixing) is simple and well established. Briefly, if a pulse of tracer gas is released in a cell of volume V_1 , then the rate of decay of tracer concentration with time is described by the equation:

$$C_{(t)} = C_0 \exp. (-N_1 t) \quad (1)$$

where $C_{(t)}$ is the tracer concentration at time t;
 C_0 is the concentration at $t = 0$;
and N_1 is the air change rate per unit time.

In practice, the air change rate, N_1 is determined from a least squares fit of $\ln C_{(t)}$ against time.

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1.2 Two cell model

The equations describing variations of tracer gas concentration in two connected cells, using two different tracer gases A and B, are far more complex than for the single cell, single tracer case. Fig. 1 shows the airflow paths for the two cell case. I'Anson *et al.*¹ have shown that the concentration of tracer gas A released in room 1, C_{A1} , at time t is given by

$$C_{A1} = \left[\frac{(F_{21} C_{O A2})/V_1 - (N_1 + Z) C_{O A1}}{Y - Z} \right] \exp Yt + \left[\frac{(Y + N_1) C_{O A1} - (F_{21} C_{O A2})/V_1}{Y - Z} \right] \exp Zt \quad (2)$$

The concentration of tracer gas A in room 2 at time t, C_{A2} , is given by

$$C_{A2} = \left[\frac{(F_{12} C_{O A1})/V_2 - (N_2 + Z) C_{O A2}}{Y - Z} \right] \exp Yt + \left[\frac{(Y + N_2) C_{O A2} - (F_{12} C_{O A1})/V_2}{Y - Z} \right] \exp Zt \quad (3)$$

where

$$2Y = -N_1 - N_2 + \sqrt{[(N_1 + N_2)^2 - 4 \{N_1 N_2 - (F_{12} F_{21})/(V_1 V_2)\}]} \quad (4)$$

and

$$2Z = -N_1 - N_2 - \sqrt{[(N_1 + N_2)^2 - 4 \{N_1 N_2 - (F_{12} F_{21})/(V_1 V_2)\}]} \quad (5)$$

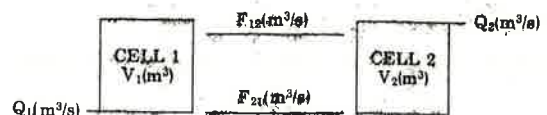


Fig. 1. Airflows between two connected cells.

If the concentration of tracer gas A is monitored in cells 1 and 2, then C_{A1} , C_{A2} , C_{OA2} , V_1 , V_2 and t are known, leaving N_1 , N_2 , F_{21} , F_{12} and Y and Z as unknowns: these are obtained using an exponential approximation known as the Prony analysis, as suggested by Hildebrand⁶. Equations (2) and (3) are rewritten as

$$C_{A1} = A \mu_1^t + B \mu_2^t \quad (6)$$

and

$$C_{A2} = C \mu_1^t + D \mu_2^t \quad (7)$$

The values of the coefficients Y and Z in Equation (3) should be real and negative. However, extraneous variables (such as changes in wind speed and wind direction during a test) can cause deviations from the expected tracer gas concentration decay and growth curves. This can result in the values of Y and Z being calculated as positive or complex, and the data set in question would have to be discarded as physically meaningless. Hildebrand⁶ discusses this occurrence, and states quite simply that this is an inherent weakness of this particular analytical technique. Where site measurements are concerned, it is clearly undesirable to be in a position where a significant proportion of the data acquired has to be rejected because of the inability of the mathematical analysis technique to deal with short term irregularities in concentration/time data. The next section re-examines the equations of conservation of tracer gas, and considers an alternative method of data analysis.

2 Alternative method for the analysis of two-directional airflows between two connected spaces

An alternative solution of the conservation of mass of tracer gas equations that describes variations in tracer gas concentrations between two connected cells is derived in Appendix 1.

Using two different tracer gases, where tracer gas A is released in cell 1 and tracer gas B is released in cell 2, the concentration of tracer gas A in cell 1, C_{A1} , at time t is given by:

$$C_{A1} = C_{OA1} \exp. (-N_1 t) + \frac{F_{21} C_{OA2}}{V_1 (N_1 - N_2)} \left[\exp. (-N_2 t) - \exp. (-N_1 t) \right] + \frac{F_{21} F_{12} C_{OA1}}{V_1 V_2 (N_2 - N_1)} \left[t \exp. (-N_1 t) + \frac{\exp. (-N_2 t) - \exp. (-N_1 t)}{N_2 - N_1} \right] \quad (8)$$

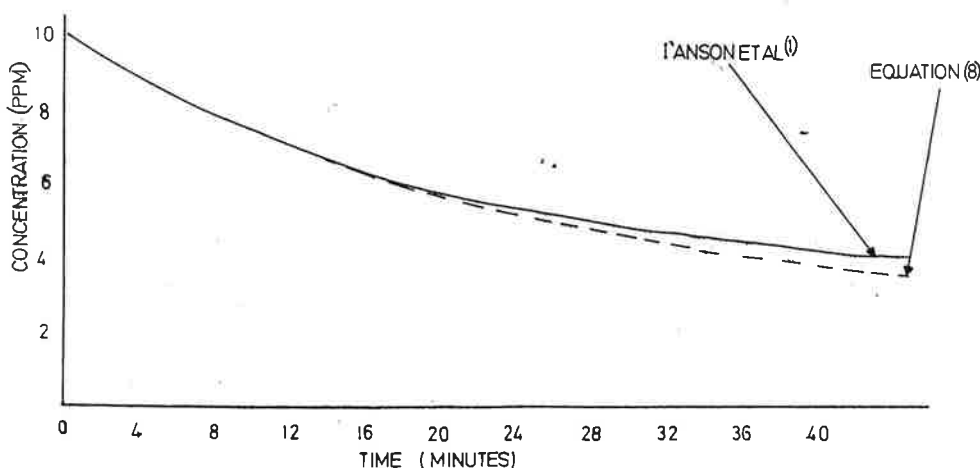


Fig. 2. $C_{A1}(t)$ variations in cell (1).

The concentration of tracer gas A in cell 2 at time t , C_{A2} , is given by:

$$C_{A2} = C_{OA2} \exp. (-N_2 t) + \frac{F_{12} C_{OA1}}{V_2 (N_2 - N_1)} \left[\exp. (-N_1 t) - \exp. (-N_2 t) \right] + \frac{F_{12} F_{21} C_{OA2}}{V_1 V_2 (N_1 - N_2)} \left[t \exp. (-N_1 t) + \frac{\exp. (-N_2 t) - \exp. (-N_1 t)}{N_2 - N_1} \right] + \frac{F_{21} F_{12} C_{OA2}}{V_1 V_2^2 (N_2 - N_1)} \left[\frac{2 \exp. (-N_2 t) - 2 \exp. (-N_1 t)}{(N_2 - N_1)^2} + \frac{t \exp. (-N_2 t) - t \exp. (-N_1 t)}{(N_2 - N_1)} \right] \quad (9)$$

The same derivation can be applied to the second tracer gas (B) released at the same time in cell 2.

Figs. 2 and 3 show the shape of Equations (8) and (9). Comparison is made with the predicted curve shapes of I'Anson *et al.*⁽¹⁾.

3 Experimental

3.1 Limitations of the existing technique

The existing technique, as used by I'Anson *et al.*¹ uses an AI 505 portable gas chromatograph, fitted with a 3 m long, 6 mm diameter 10 per cent squalane separation column, used in conjunction with an electron capture detector. An internal diaphragm pump in the instrument draws air from the cell under test. A six port sampling valve isolates a fixed volume of the air-tracer gas mixture: this is carried through the separation column using argon as a carrier gas. In the column each gas present is absorbed and desorbed at different rates by the squalane packing, causing the component gases to arrive at the other end of the column at different times. The separated gas mixture passes through the electron capture detector where the presence of an electron absorbing gas causes a drop in the standing current generated in the detector cell. The drop in standing current can be measured and related to the concentration of this gas.

Tracer gases are introduced into the test space in one of two ways. The first being to inject the tracer gas remotely using a gas tight syringe, via a septum port into a polythene tube which is flushed with argon. Alternatively, tracer gas may be injected manually from the gas cylinder

in the appropriate space by briefly opening the cylinder valve.

After injection the tracer gases are mixed using oscillating desk fans, sampling of the air tracer gas mixture being achieved by drawing air through polythene tubes (8 mm O.D.) from each space under test.

This piece of equipment has a sampling interval, for the two cell case, of 1 minute. In order to reduce errors in the values of airflows derived by the new simplified analysis, sufficient data points must be acquired within 15 minutes of the start of the test, that is, before recirculation terms become significant. On the basis of 10 data points per cell per test, a sampling interval of 45 seconds or less is required. Clearly, some improvement in the rate of sampling of the existing gas chromatograph was needed. Two ways of improving the sampling interval, whilst still retaining the same type of basic unit, were considered.

The first option was to duplicate the existing equipment and to sample alternatively with each. This was rejected for three reasons: first, the effective reduction in portability caused by having to use twice as much equipment; secondly, the expense involved; and thirdly, but from an experimental viewpoint most importantly, the uncertainty of obtaining two AI 505 gas chromatographs with identical output characteristics. For example, the perfect matching of the electron capture detector amplifier board is unlikely and differences of up to 40 per cent have been observed.

The option selected was to use two gas chromatographic separation columns of identical packing and length attached in parallel to one gas chromatograph unit. Sampling valves would be arranged so that samples passing through each column would be passed alternately to the electron capture detector. A faster sampling rate is achieved by virtue of the fact that the 'dead time' associated with waiting for a sample to pass through a single column is eliminated. Such an arrangement has two very attractive advantages: first, the portability of the original apparatus would be hardly changed; and secondly, the cost of such a system is considerably less than duplicating the AI 505 unit. The major technical difficulty envisaged was ensuring that the two separation columns behaved in an identical manner, though, as events transpired, this was not as big a problem as originally feared.

3.2 Design of the new system

For 2 zone work, it is sufficient to be able to detect 2 Freons on a gas chromatograph, in this case Freon 12 and Freon

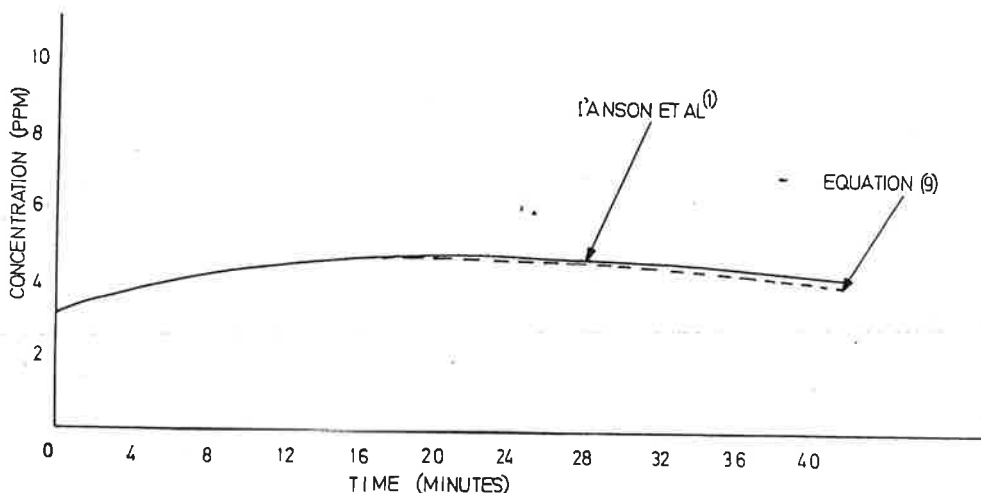


Fig. 3. $CA_2(t)$ variations in cell (2).

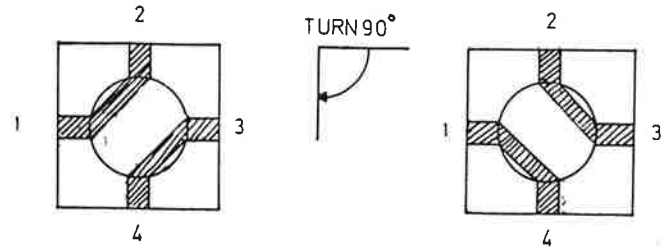


Fig. 4. 4-port valve.

114. Previous experience with single column operation has shown that the best compromise between throughput time and peak resolution is given by a column packing of 10 per cent squalane on a CNAW support, with a column length of 3 m and diameter 6 mm.

It is essential that the columns are both kept under a trickle of gas during operation. If the gas stream to either column were to be interrupted during operation, then gas starvation and retention of atmospheric contaminants would take place, with resulting deleterious effects on system stability. Therefore, the sampling valve system to be used had to provide for carrier gas trickle over the column not directly connected to the electron capture detector. Such a flow pattern could have been achieved using an arrangement of simple 2-way valves. However, for ease of operation, two four-port zero dead volume valves, as manufactured by Whitey Valves Inc., were chosen. By a single 90° turning action, two inputs can be directed via either of two outlets (Fig. 4).

A schematic diagram of the new system is shown in Fig. 5. The position of valve 1 determines the column to which the gas sample is diverted: the sampling valve used is the one originally supplied with the AI 505 model. The needle valves located after the separation columns are used to equalise the pressures in both legs of the system, ECD output being sensitive to operating pressure. The position of valve 2 determines whether the gas stream from a separation column is sent to the electron capture detector or else to exhaust. Preliminary tests showed that the resistance of the ECD was significantly higher than that of the exhaust section of the system: this affects baseline stability when columns are switched over. To overcome this, a needle valve is situated in the exhaust line, enabling its resistance to be matched to that of the ECD.

The pattern of valve switching which would be used during a ventilation test in which a 30 second sampling interval was employed is summarised in Table 1.

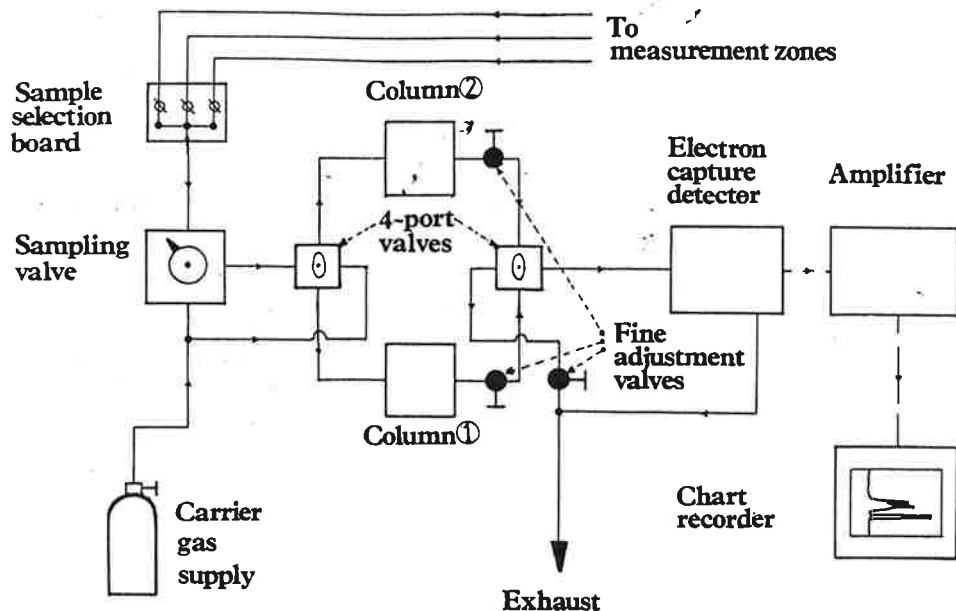


Fig. 5. Schematic layout of rapid sampling system.

Table 1. 4-port valve switching sequence: 30 second sampling.

Time (sec)	Action	Position of Valve 1	Position of Valve 2	System Status
0-3	Inject Sample	a	b	Sample directed to column 1.
12	Switch 1	b	b	Sample going through column 1: column 2 on line ready for next sample.
23	Switch 2	b	a	ECD on line to column 1. Ready to receive sample.
30-33	Inject Sample	b	a	First sample shows an ECD output: second sample column 2.
42	Switch 1	a	a	First sample still being output: second sample going through column 2: column 1 on line ready for next sample.
52	Switch 2	a	b	First sample finished with: ECD on line ready to receive second sample.
60-63	Inject Sample	a	b	Second sample shows as ECD output: third sample directed to column 1.

3.3 Problems encountered during commissioning

After initial difficulties involving pressure imbalances within the system had been resolved, the single most potentially serious problem would be variations in response between the two separation columns. With this in mind, it was decided to adopt the following three conditions as standard operating procedure in an attempt to minimise potential difficulties:

- (1) Columns to be bought from the same manufacturer, and to be packed during the same production run from the same batch of packing material.
- (2) Both columns to be baked in parallel in an oven at 100°C for 12 hours, with the purging gas to be drawn from the same cylinder.
- (3) When not in use, both columns to be kept under a blanket of argon in parallel, with the purging gas to be drawn from the same cylinder.

With these conditions imposed, laboratory tests showed that against a background tracer gas concentration of 40 per cent of full scale deflection, the maximum difference in response between any pair of columns used was not greater than 2.5 per cent. Whilst this was a reasonable starting point, it was clearly highly desirable to reduce any potential systematic error to the absolute minimum.

Attention was then turned to the effect of column operating temperature. At room temperature, small fluctuations in temperature can have significant effects on the

performance of a column, both in terms of sample throughput time for a given carrier gas pressure and, more importantly, baseline stability. If such fluctuations and variations could be eliminated, then it was conceivable that column performances would become more closely matched.

To investigate this possibility, a small thermostatically controlled water bath/stirrer unit was purchased, with intention of holding both columns at a constant operating temperature, $\pm 0.1^\circ\text{C}$. Clearly, the holding temperature had to be above the likely range of ambient temperatures which could be encountered; to this end, the water bath thermostat was set at a nominal 30°C (86°F). At this temperature, against a background tracer gas concentration of 40 per cent of full scale deflection, the maximum difference in response between any pair of columns used was not greater than 0.5 per cent; furthermore, the higher operating temperature effectively eliminated baseline drift problems previously encountered with the system at ambient temperature operating conditions.

4 Results and discussion

The specimen result presented here shows the flow of air between the upstairs and downstairs of a dwelling. Freon 114 was released downstairs, and Freon 12 upstairs. Fig. 6 shows the decay of Freon 114 downstairs and its growth upstairs; Fig. 7 shows the decay of Freon 12 upstairs and its growth downstairs. The analysis of this data, using the curve fitting technique described in Appendix 1 gives the

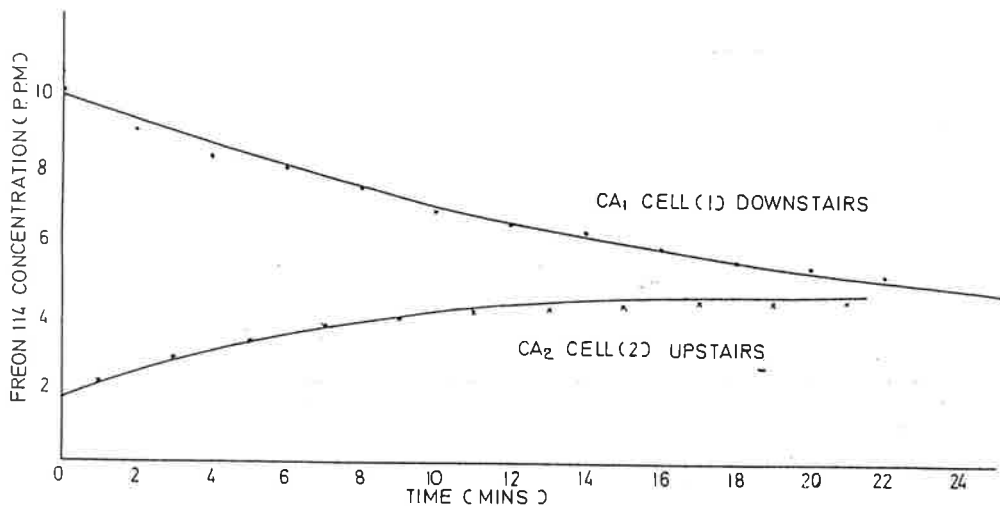


Fig. 6. Freon 114 released downstairs.

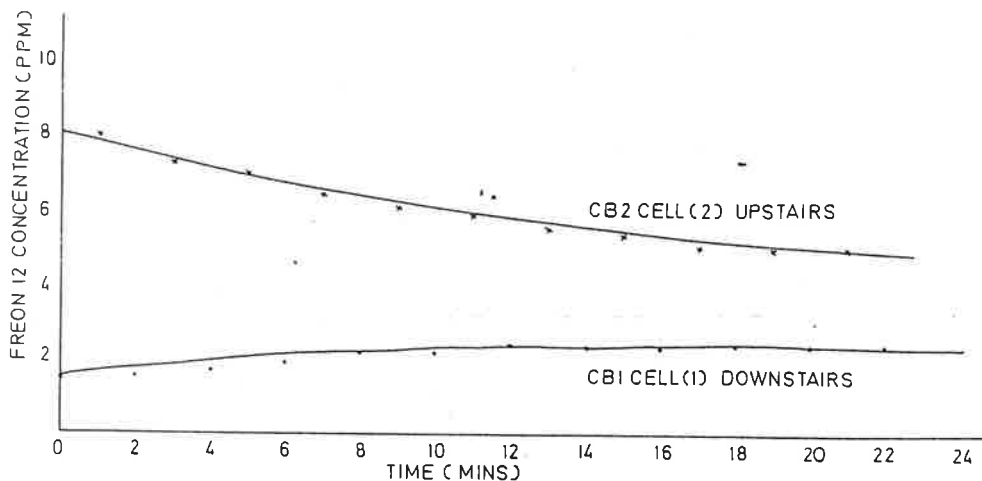


Fig. 7. Freon 12 released upstairs.

following values of ventilation and airflow rates: $N_1 = 2.5$ ach, $F_{21} = 230$ m³/h; $N_2 = 2.1$ ach, $F_{12} = 125$ m³/h. The uncertainty in these values is of the order of ± 10 per cent, based on a series of validation tests carried out in two interconnected environmental chambers^{7,8}.

The simplified analytical solution enables estimates of air change rates and intercell airflows to be calculated directly from site measurements of the variations of tracer gas concentrations with time. The new method can cope more readily with random measurement errors than the Prony method used previously¹. The rapid sampling system shortens the length of tests, and hence minimises the effects of extraneous variables (e.g. wind speed and direction) which could otherwise cause significant variations in site data.

5 Conclusions

The new improved trace gas analysis system, used in conjunction with the simplified mathematical analysis for the airflow between two interconnected zones, is a useful tool for measuring airflows and ventilation rates in a short time period, enabling errors due to the effects of extraneous variables on site data to be minimised. Currently, the technique and analysis are being extended to the cases of three and four interconnected cells.

The usefulness of this technique with regard to ventilation and air movement measurements in large single cells (i.e. industrial premises) is currently being evaluated.

The system is also being fully automated, both in terms of valve control and data acquisition. An Apple IIe micro-computer, in conjunction with analogue to digital and digital to analogue conversion modules, is being used to fulfil both functions. A suite of data analysis and data manipulation programs is also being developed; again this will be used on the Apple IIe.

APPENDIX 1

Analysis of two cell airflows N_1 , N_2 , F_{12} and F_{21} between two connected spaces

Considering Fig. 1, if tracer gas A is released in cell 1 and allowed to mix with the air, some of it will be carried to cell 2, where it will also mix with the air, after which some may be returned to cell 1. Let us define $t = 0$ (seconds) as the time when initial mixing is complete: the concentrations of tracer gas A in cells 1 and 2 are given by C_{OA1} and C_{OA2} respectively. Now the rate at which tracer gas A is entering cell 1 is given by $F_{21} C_{A2}$ and the rate at which it is leaving by $F_{12} C_{A1} + Q_1 C_{A1}$; hence, the rate of decrease of volume of tracer gas A in cell 1 at time t is given by.

$$-V_1 \frac{dC_{A1}}{dt} = C_{A1}(F_{12} + Q_1) - F_{21} C_{A2} \quad (1)$$

Similarly, the rate of decrease of volume of tracer A in cell 2 is given by:

$$-V_2 \frac{dC_{A2}}{dt} = C_{A2}(F_{21} + Q_2) - F_{12} C_{A1} \quad (2)$$

Since

$$N_1 = \frac{F_{12} + Q_1}{V_1} \text{ and } N_2 = \frac{F_{21} + Q_2}{V_2}$$

Equations (8) and (9) become:

$$\frac{dC_{A1}}{dt} + N_1 C_{A1} = \frac{F_{21}}{V_1} C_{A2} \quad (3)$$

and

$$\frac{dC_{A2}}{dt} + N_2 C_{A2} = \frac{F_{12}}{V_2} C_{A1} \quad (4)$$

Using exp. ($N_1 t$) as the integrating factor for Equation (3);

$$C_{A1} \exp. (N_1 t) = \int \frac{F_{21} C_{A2}}{V_1} \exp. (N_1 t) dt + A \quad (5)$$

and using exp. ($N_2 t$) as the integrating factor for Equation (4);

$$C_{A2} \exp. (N_2 t) = \int \frac{F_{12} C_{A1}}{V_2} \exp. (N_2 t) dt + A \quad (6)$$

For Equation (5) to be solved, the variation with time of C_{A2} must be known or approximated. If it is assumed that, initially, no recirculation of tracer gas A occurs (i.e. $F_{21} = 0$) then, from Dick's equations², C_{A2} is given by:

$$C_{A2} = C_{OA2} \exp. (-N_2 t) + \frac{F_{12} C_{OA1}}{V_2 (N_2 - N_1)} \left[\exp. (-N_1 t) - \exp. (-N_2 t) \right] \quad (7)$$

Substituting for C_{A2} in (5) and solving:

$$C_{A1} = C_{OA1} \exp. (-N_1 t) + \frac{F_{21} C_{OA2}}{V_1 (N_1 - N_2)} \left[\exp. (-N_2 t) - \exp. (-N_1 t) \right] + \frac{F_{21} F_{12} C_{OA1}}{V_1 V_2 (N_2 - N_1)} \left[t \exp. (-N_1 t) + \frac{\exp. (-N_2 t) - \exp. (-N_1 t)}{N_2 - N_1} \right] \quad (8)$$

Similarly, Equation (6) can be solved by substitution for C_{A1} to give:

$$C_{A2} = C_{OA2} \exp. (-N_2 t) + \frac{F_{12} C_{OA1}}{V_2 (N_2 - N_1)} \left[\exp. (-N_1 t) - \exp. (-N_2 t) \right] + \frac{F_{12} F_{21} C_{OA2}}{V_1 V_2 (N_1 - N_2)} \left[t \exp. (-N_2 t) + \frac{\exp. (-N_2 t) - \exp. (-N_1 t)}{N_2 - N_1} \right] + \frac{F_{21} F_{12}^2 C_{OA1}}{V_1 V_2^2 (N_2 - N_1)} \left[\frac{2 \exp. (-N_2 t) - 2 \exp. (-N_1 t)}{(N_2 - N_1)^2} + \frac{t \exp. (-N_2 t) + t \exp. (-N_1 t)}{N_2 - N_1} \right] \quad (9)$$

The same derivation can also be applied to a second tracer gas (B) released at the same time in cell 2.

If the concentrations of tracer gas A are monitored in cells 1 and 2, then C_{A1} , C_{A2} , t , V_1 and V_2 are known, leaving N_1 , N_2 , F_{12} , C_{OA1} and C_{OA2} as unknowns. By using the following curve fitting technique on sets of experimental values of C_{A1} , C_{A2} and t , these unknowns can be calculated.

The technique discussed here assumes the effects of recirculation of tracer gas between connected cells is time dependent, the contribution of recirculated tracer gas being small for time (t) < 20 minutes. Consequently Equations (8) and (9) for $C_{(t)}$ can be simplified to:

Considering Cell 1

Gas A

$$\tilde{C}_{A1} = C_{OA1} (1 - A N_1) + \frac{F_{21} C_{OA2}}{V_1 (N_1 - N_2)} \quad (10)$$

[exp. (- $N_2 t$) - exp. (- $N_1 t$)]

Gas B

$$\tilde{C}_{B1} = C_{OB1} (1 - A N_1) + \frac{F_{21} C_{OB2}}{V_1 (N_1 - N_2)} \quad (11)$$

[exp. (- $N_2 t$) - exp. (- $N_1 t$)]

Considering Cell 2

Gas A

$$\tilde{C}_{A2} = C_{OA2} (1 - B N_2) + \frac{F_{12} C_{OA1}}{V_2 (N_2 - N_1)} \quad (12)$$

[exp. (- $N_1 t$) - exp. (- $N_2 t$)]

Gas B

$$\tilde{C}_{B2} = C_{OB2} (1 - B N_2) + \frac{F_{12} C_{OB1}}{V_2 (N_2 - N_1)} \quad (13)$$

[exp. (- $N_1 t$) - exp. (- $N_2 t$)]

where

$$\tilde{C}_{(t)} = \left[\int_{t=0}^{t=t} C_{(t)} dt \right] / \Delta t$$

the integral is evaluated using numerical integration of site $C_{(t)}$ data points.

(1 - $A N_1$) is a Maclaurin series expansion of exp. (- $N_1 t$)

$$A = -t + \frac{N_1^2 t^2}{2!} - \frac{N_1^3 t^3}{3!} + \frac{N_1^4 t^4}{4!} - \frac{N_1^5 t^5}{5!}$$

(1 - $B N_2$) is a Maclaurin series expansion of exp. (- $N_2 t$)

$$B = -t + \frac{N_2^2 t^2}{2!} - \frac{N_2^3 t^3}{3!} + \frac{N_2^4 t^4}{4!} - \frac{N_2^5 t^5}{5!}$$

N_1 , N_2 are 'first-order' estimates of N_1 and N_2 , found from taking the $C_{(t)}$, time (t) for C_{A1} and C_{B2} data points.

Equations (10) and (11) can be solved using numerical iteration for N_1 , F_{21} this gives:

$$N_1 = \frac{1}{A} - \frac{\tilde{C}_{A1}}{C_{OA1} A} + \frac{F_{21} C_{OA2}}{C_{OA1} A V_1 (N_1 - N_2)} \quad (14)$$

[exp. (- $N_2 t$) - exp. (- $N_1 t$)]

$$F_{21} = \frac{V_1 (N_1 - N_2)}{C_{OB2}} [\text{exp.} (- N_2 t) - \text{exp.} (- N_1 t)] \quad (15)$$

[$\tilde{C}_{B1} - C_{OB1} (1 - A N_1)$]

Similarly Equations (13) and (12) give

$$N_2 = \frac{1}{B} - \frac{\tilde{C}_{B2}}{C_{OB2} B} + \frac{F_{12} C_{OB1}}{V_2 (N_2 - N_1) C_{OB2} B} \quad (16)$$

[exp. (- $N_1 t$) - exp. (- $N_2 t$)]

$$F_{12} = \frac{V_2 (N_2 - N_1)}{C_{OA1}} [\text{exp.} (- N_1 t) - \text{exp.} (- N_2 t)] \quad (17)$$

[$\tilde{C}_{A2} - C_{OA2} (1 - B N_2)$]

The iteration generally converges in less than ten steps.

The calculated values of N_1 , N_2 , F_{12} and F_{21} can be used in Equations (8) and (9) to enable comparison between theoretical curve shapes and the site data points.

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