BASELINE CARBON DIOXIDE DATA SELECTION

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ABSTRACT

Methods of obtaining suitable selection criteria for carbon dioxide monitoring at baseline stations, are discussed. The meaning of "baseline" is defined and a number of chemical species and physical parameters assessed for suitability as baseline criteria.
A description is given of the methods employed in determining suitable parameters for data selection at the Cape Grim baseline station and an assessment made of their success.

Baseline conditions :

REPRESENTATIVE OF LARGE SPACE SCALES

with

SMALL SURFACE EXCHANGE RATES (sources and sinks)

and

STRONG VERTICAL MIXING

Parameters suitable for use as selection criteria:

WIND DIRECTION

WIND SPEED

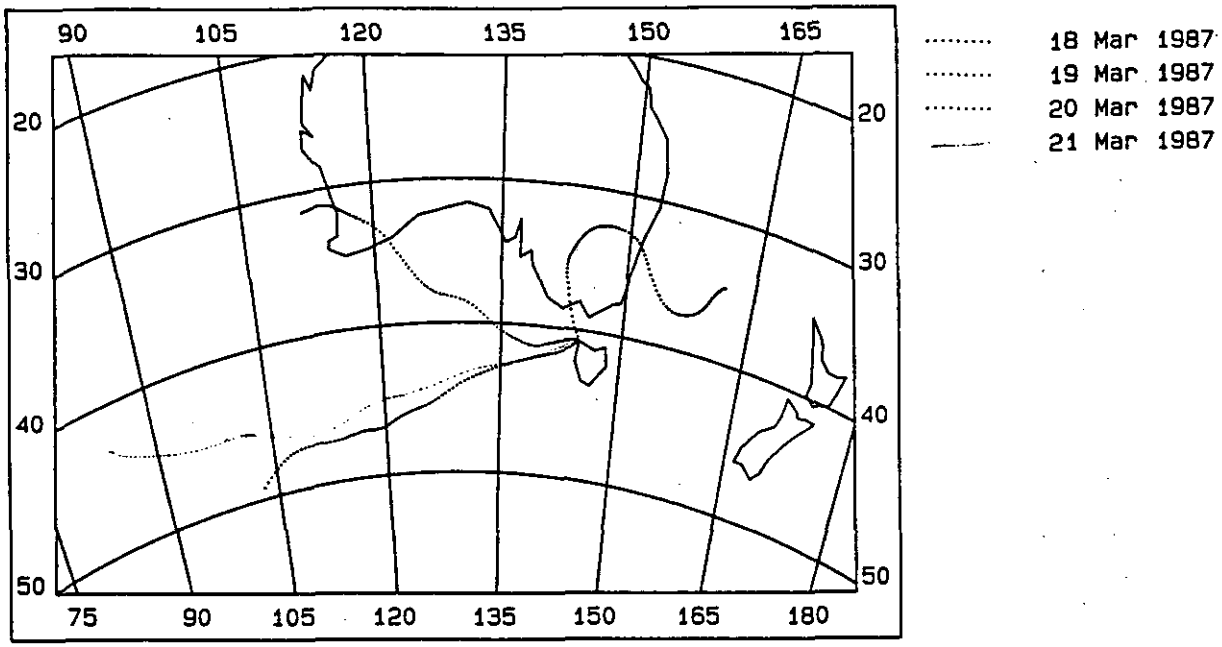
CONSISTENCY OF CO_2

PARTICLE COUNT

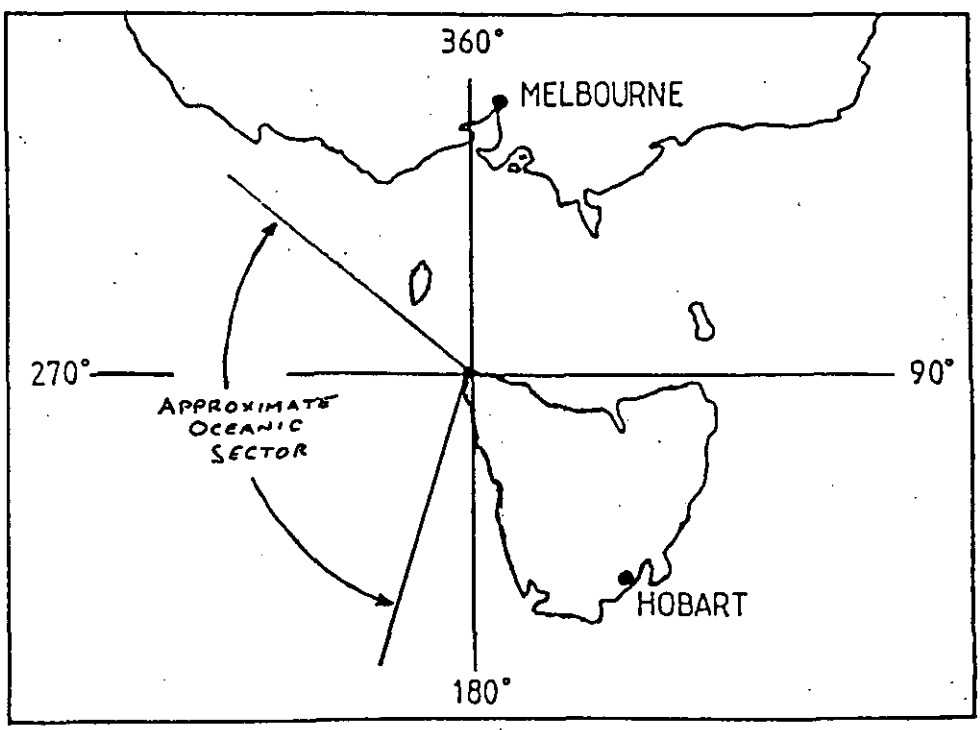
RADON

CARBON 14 (^{14}C)

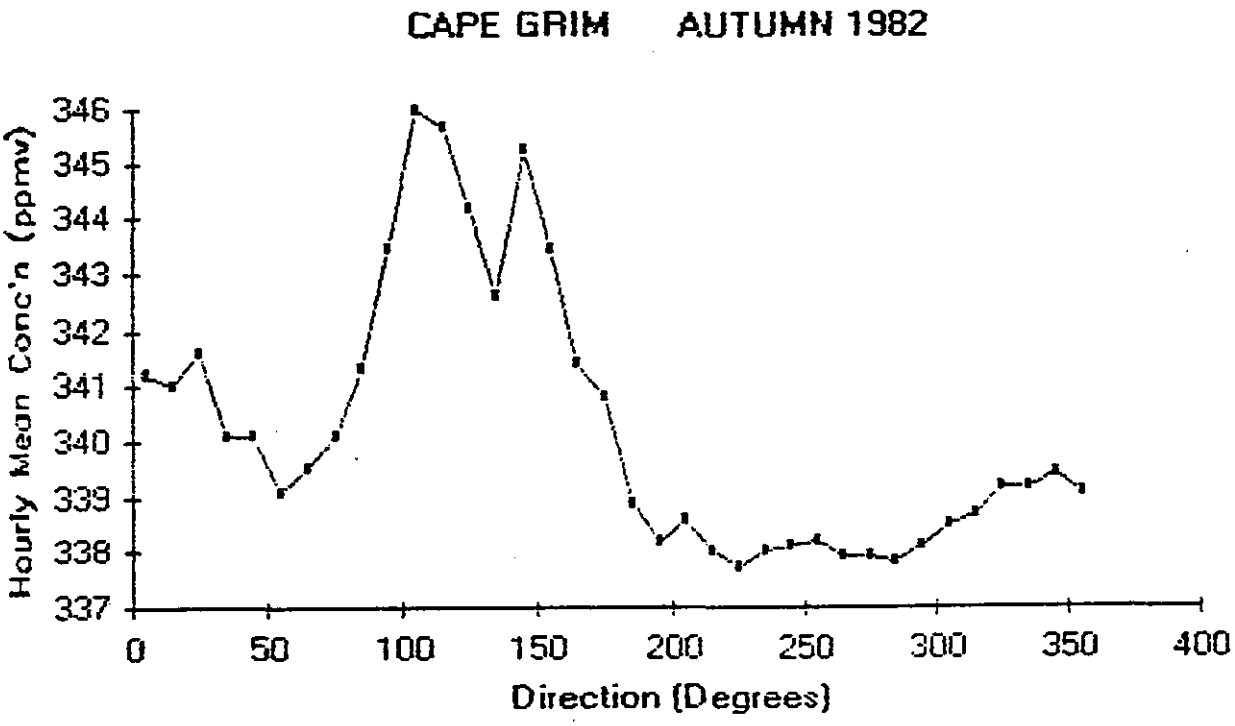
AIR MASS TRAJECTORIES



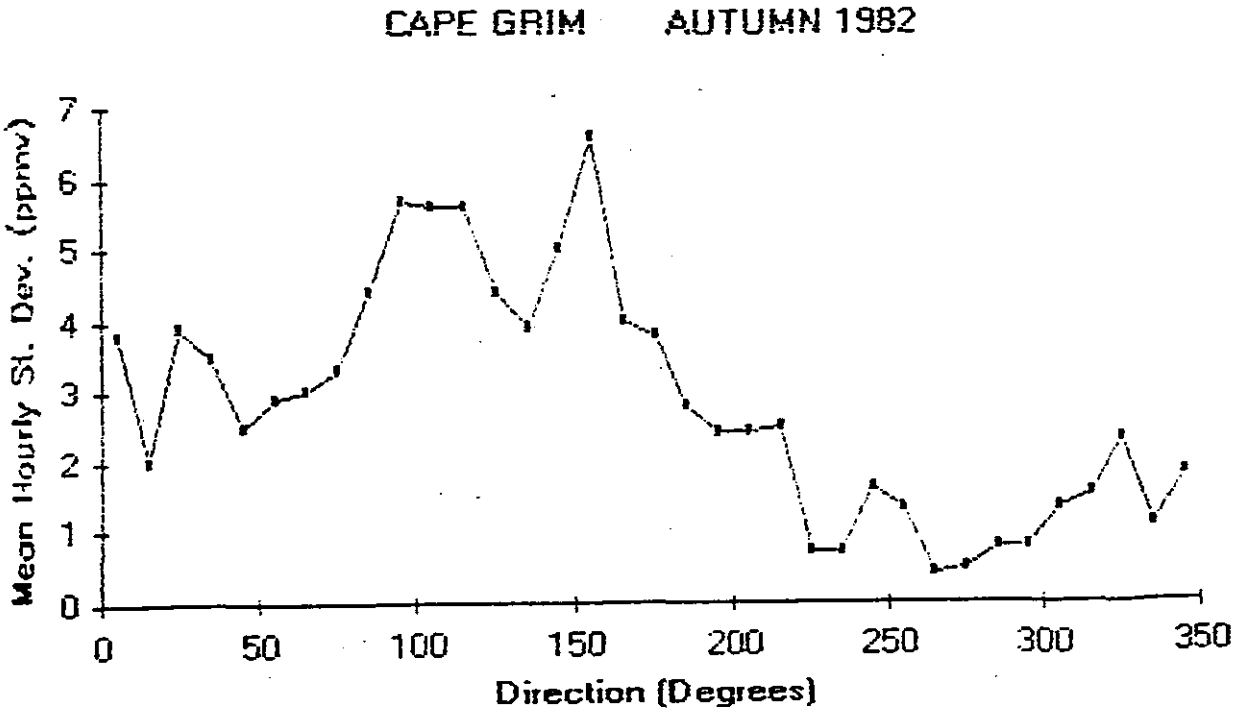
Typical air mass trajectories to Cape Grim baseline station.



Approximate oceanic (baseline) sector at Cape Grim.



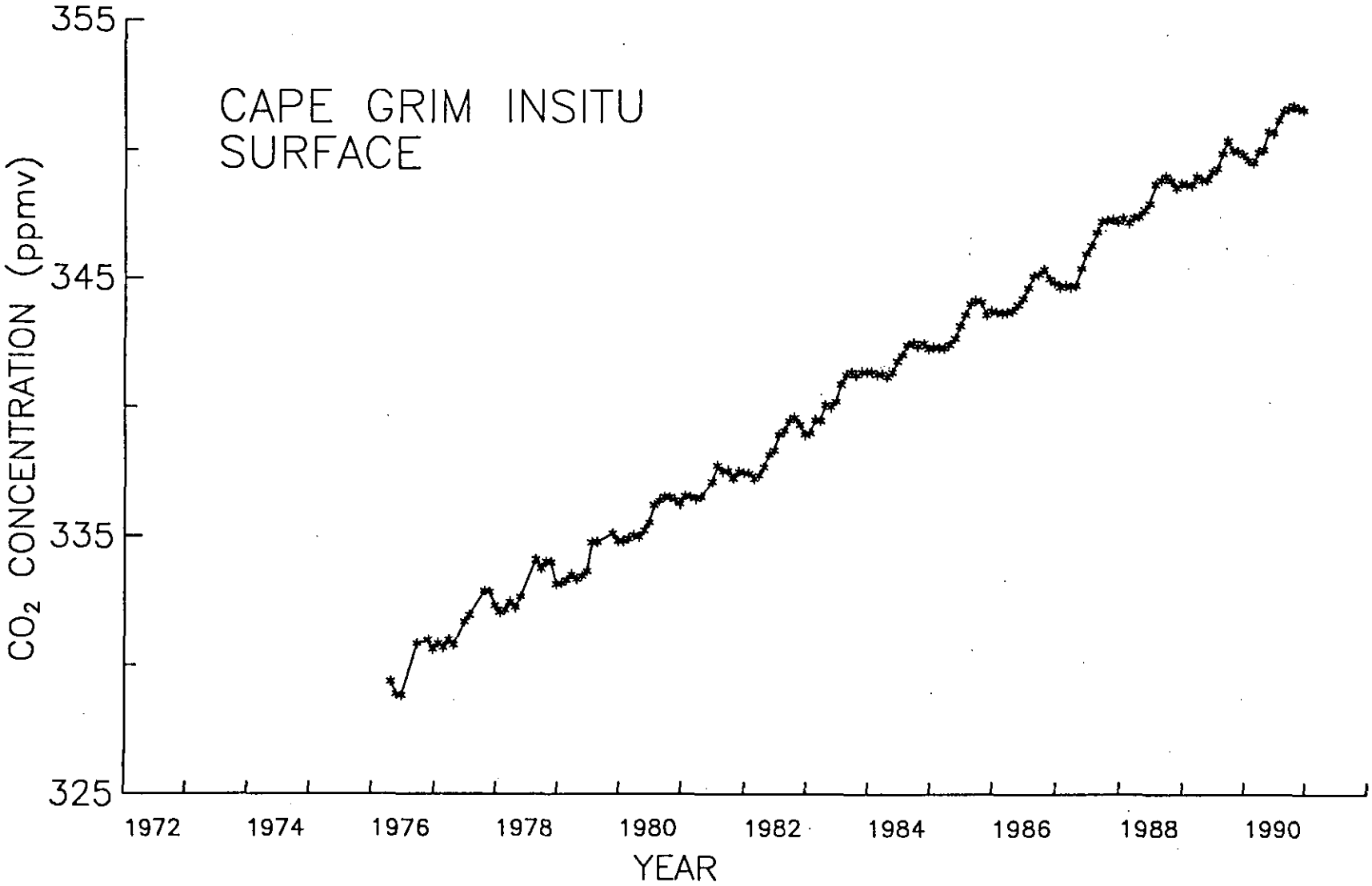
Mean hourly CO2 concentrations in 10 degree sectors for Autumn, 1982 at Cape Grim.



Mean standard deviations on hourly means in 10 degree sectors for Autumn, 1982 at Cape Grim.

Influence of selection criteria applied to hourly mean CO_2 data at Cape Grim for March 1982

SELECTION CRITERIA	MEAN CO_2 (ppmv)	S.D. (ppmv)	NO. OF HOURS	NO. OF DAYS
Total data	340.12	2.87	726	31
Wind direction 190^o - 280^o	337.92	1.76	253	17
Wind speed >5 ms^{-1}	340.39	3.60	592	31
CO_2 consistency				
5 hr; < +/- 0.3 ppmv	337.50	0.59	227	19
5 hr; < +/- 0.5 ppmv	337.98	1.16	298	25
5 hr; < +/- 1.0 ppmv	338.37	1.33	457	29
10 hr; < +/- 0.3 ppmv	337.31	0.45	157	12
10 hr; < +/- 0.5 ppmv	337.47	0.58	191	15
10 hr; < +/- 1.0 ppmv	338.02	1.24	316	24
W/D: 190^o-280^o W/S: > 5 ms^{-1}	337.34	0.42	205	14
W/D: 190^o-280^o W/S: > 5 ms^{-1} Cons: 5 hr; < +/- 0.3 ppmv	337.19	0.32	172	12
W/D: 190^o-280^o W/S: > 5 ms^{-1} Cons: 10 hr; < +/- 0.3 ppmv	337.12	0.24	123	8
W/D: 190^o-280^o Cons: 5 hr; < +/- 0.3 ppmv	337.22	0.36	180	13
W/D: 190^o-280^o Cons: 10 hr; < +/- 0.3 ppmv	337.12	0.24	123	8


CAPE GRIM INSITU
SURFACE
355
345
335
325
CO_2 CONCENTRATION (ppmv)
1972
1974
1976
1978
1980
1982
1984
1986
1988
1990
YEAR

THE ROLE OF RADON AND RADON DAUGHTERS IN AIR MASS CHARACTERISATION AT BASELINE ATMOSPHERIC OBSERVATORIES

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ABSTRACT

The problem of baseline selection is examined at two two contrasting sites: Cape Grim, with a very strong close source of pollution but only slight influences from distant sources, and Mauna Loa Observatory, with only weak local sources, but periods of weeks when anthropogenic pollution is transported rapidly from Asia. In both cases, radon can be used as an indicator of land contact. Examples of the relationship between radon and trace gas concentrations are given for each site, demonstrating the importance of radon in understanding the previous history of air samples.

Radon measurements at baseline concentrations take the order of an hour, which is too slow for control of conditional samplers. Radon daughter detectors can be made to work faster, but provide only a poor estimate of radon concentration. A recently developed instrument at Cape Grim demonstrates that radon estimates suitable for baseline selection can be made within 20 minutes by combining radon daughter with CN concentration. The weather conditions at Cape Grim are such that this permits elimination of more than 90% of samples with above threshold radon concentrations in what would otherwise be considered baseline air.

1. Introduction

The primary function of baseline atmospheric observatories is to monitor long term trends in concentrations of trace constituents of the atmosphere. In selecting air samples it is especially important to separate air masses which are baseline from those which have been subject to recent pollution. The definition of "baseline" is somewhat species-dependent, but it is generally accepted that air samples recently perturbed by passage over large land masses, cities etc. cannot be accepted as "baseline".

Gras et al (1992) showed that a combination of three types of data are necessary for establishing a reasonably objective method for baseline selection:
a). Meteorological data can be used establish the recent path of air. Within a few hundred km of a pollution source wind direction can often define baseline conditions. Beyond that, trajectory calculations provide a good general idea of the path the air has taken, but there are three important limitations: the information is not immediately available, so that it cannot be used to operate conditional samplers; the timing accuracy is generally no better than a few hours, so the data cannot be used to select pollution episodes during periods of change; and even though air may have passed over land, it may not actually have mixed with air from the surface.

b). Condensation nucleus (CN) concentration is a useful indicator of pollution by burning or fuel use such as in industry or transport. A key feature of CN is that they can be measured essentially instantaneously, and so are ideal for control of samplers. However, when the source of pollution is more than a few hundred km from the observatory, CN loss and production processes in transit reduce the effectiveness of CN as a tracer for pollution.
c). Radon is an excellent tracer for air masses which have passed over land within the last ten or so days (Larson 1973, Wilkness et al 1973, Lambert et al 1970, Whittlestone 1985b, Downey et al 1990, Pereira et al 1988, H.E.Moore et al 1974). This is because radon is an inert gas unaffected by chemical interactions in transit, and its emanation rate from land is about one hundred times the rate from the ocean (Wilkening et al, 1975). Its half-life of 3.8 days ensures that it does not build up in the atmosphere. Limitations in the use of radon are the slow response time (1 to 2 hours) of detectors currently employed and the requirement of several km path over land to produce a measurable increase the radon concentration.

The two laboratories from which data for the present study are taken (see Figure 1) are at Cape Grim (CG) (40.7° S, 144.7°E) on Tasmania and Mauna Loa Observatory (MLO) (19.5°S, 155.6°W) on Hawaii. CG has prolonged periods of unpolluted westerly winds, but frequently experiences inflow from Tasmania to the south east and the Australian continent to the north. MLO is located on the NNE slope of the Mauna Loa volcano at an altitude of 3400 m. It has larger towns close to the observatory than CG, but beyond them are several thousand km to the nearest significant land mass. In addition, the local meteorology of MLO leads to air being sampled sometimes from the boundary layer and sometimes from the free troposphere. Transport processes bringing polluted air to these observatories range from very local small-scale to large-scale convective transport in a distant land followed by fast long-range transport in the free troposphere.

Methane and radon concentrations at MLO are presented to demonstrate the strong influence of fast long range transport on air quality. Measurements at CG of ozone and radon show that correlations of species with different source and transport properties can improve our understanding of air samples. In all the measurements, a meteorological baseline criterion is used. As recommended by Gras et al, conditions were deemed to be baseline only when all the baseline meteorological conditions had persisted for at least two hours.

Given the importance of radon in selecting air subject to recent pollution, it would clearly be desirable to be able to control samplers according to the prevailing radon concentration. Data are presented from an instrument recently installed at CG, to achieve this goal.

2. Siting of Observatories and their Local Meteorology

Local influences are small at CG. It is surrounded by sea except for the S to SE sector, which is low, flat and well covered by vegetation. Land/sea breeze effects are generally small, and vestigial in strong westerly winds. Only a few percent of winds come from the

direction of Tasmania (Downey et al 1990). However, an appreciable fraction of the winds are from the Australian continent which subtends a large angle at CG. There are therefore many occasions when polluted air reaches the station after medium length paths (a few hundred to a thousand km) over the ocean.

Hawaii is so far from large land masses that wind direction is a very poor guide to the origin of sampled air, in either the boundary layer or the free troposphere. Weather systems which prevail in the Pacific ocean generally result in strong separation of the two layers by a temperature inversion at about 2000m, and transit times from continents of the order of weeks. Periodically one or other layer may receive continental air after only a few days which can lead to samples not representative of the baseline at MLO. Transport of dust and radon during such periods from continental Asia to Hawaii has been well documented (Moore et al 1974, Darzi et al 1982,Blank et al 1985). At other times the synoptic air flows, usually the easterly trade winds, are accompanied by turbulent mixing between the layers.

At MLO synoptic air flows are superimposed on diurnal convective flows which predominate under calm conditions with only light cloud cover. Then the daytime wind is upslope (northerly) and air at the observatory comes from close to sea level, passing low over the intervening land. The air is moist, its trace gas concentrations perturbed by its proximity to land, and it may have been polluted by passage over a town. At night dry air is brought down-slope from the free troposphere. There is only a short passage over barren recent lava which has minimal influence on the air quality.

When there is cloud cover or a strong synoptic flow, air sampled is virtually unaffected by local land and is representative of the station altitude. Whilst this may be dry free tropospheric air, on some occasions it is a mixture of boundary layer and free tropospheric air, resulting in medium moisture content. However, it is rare under these conditions to observe the very high moisture levels typical of calm upslope conditions.

3. Instrumentation

At Cape Grim CN are measured with a TSI model 3020 CN counter. It is referenced to the CSIRO standard Nolan/Pollak counter, the calibration of which is discussed by Gras (1984).

The radon detectors are modelled on that described by Whittlestone (1985a). The sensitivities of the detectors are 0.23 and 0.19 counts s^{-1} for concentrations of 1 Bq m^{-3} at CG and MLO respectively. A 30% statistical error from a one hour count is obtained at concentrations of 10 and 15 mBq m^{-3}. Background count rates are equivalent to 10 and 20 mBq m^{-3} respectively. Response times are 90 minutes to 50% of maximum. The CG and MLO detectors are calibrated automatically at the start of each month by passing radon from a source through the detector for four hours (Whittlestone et al, 1991).

The Cape Grim Automatic Radon Daughter Detector (GADD) has described in detail elsewhere (Whittlestone 1992). It employs an automatic filter changer, and operates on a 20 minute cycle. Air is drawn through the filter at 270 L/min. The decay of radon daughters trapped on the filter is monitored by an alpha particle detector whose output is fed into a computer. At the end of the 20 minute cycle, the integrated count is a measure of the radon daughter concentration.

Methane was measured by a gas chromatograph with flame ionisation detection and a computer-based integrator. It is described in detail by Dlugokencky et al (1991). Details of the ozone measurement system at Cape Grim are given by Elsworth et al (1984).

4. Methane and radon measurements at MLO.

Methane has a lifetime in the atmosphere of about 12 years, governed mainly by destruction by the OH radical. The major sources of methane are land-based, with rice cultivation and ruminant animals in Asia the predominant sources upwind of MLO. Methane concentration at MLO exhibits a strong seasonal cycle superimposed on a steady increase. Harris et al (1992) have shown that elevated methane correlates well with periods when air trajectories indicate recent passage of the sampled air over Asia. In this study the time scales were synoptic to seasonal, shorter time scales being precluded by the inherent timing imprecision of trajectory analysis. With radon measurements as a guide to continental contact, it is possible to study the variability of methane concentrations on a time-scale of an hour. This discussion will focus just on the seasonal variation of methane hourly averages selected according to the prevailing radon concentration.

Methane concentrations were selected from periods when meteorological conditions precluded significant influence from the island of Hawaii. These data were divided into sets corresponding to several radon concentration ranges, the lowest of which was the zero to 100 mBq m^{-3} band suggested by Gras et al. to be a reasonable "baseline" criterion. The seasonal dependence of each set is shown in Figure 2. Also shown is the annual trend based observed over five years.

The methane concentrations in the lowest radon band are seen to increase by about 20 ppbv from September through February compared to the other months. This agrees well in both time and magnitude with the model of Fung et al (1991), which estimates an average annual variation at the MLO latitude of 25 ppbv with a high period from September through April. The seasonal dependences of methane from samples with higher radon concentrations are clearly strongly divergent from the prediction, both in magnitude and timing.

This supports the use of radon as a selection criterion for any gas influenced by passage over land. Discrepancies between a model and measurements can be assessed by examining sub-sets of the data. In the case of the model of Fung et al (1991), the implication of the radon selection process is that the model estimates long term averages, and not the transient effects of episodes of fast transport from Asia to

MLO. The present data justify the use of the baseline threshold of 100 mBq m^{-3} at MLO, as suggested by Gras et al (1992).

5. Ozone and radon measurements at Cape Grim

Ozone is a much more reactive gas than methane. Its sources at CG are:
i) mixing down from the stratosphere;
ii) photochemical production in the troposphere;
iii) production in plumes from distant urban centres.
The influence of land is largely destruction of ozone by contact with vegetation. Emmissions from urban and industrial complexes produce ozone by photochemical processes, so the effect of passage over land can be either a loss or an enhancement, depending on the exact trajectory.

The primary question of concern here is whether the presence of air of recent continental origin in baseline samples is likely to affect the ozone record. A very broad view was required to try to find order in a very complex data set. The approach taken was to look for general trends in baseline and non-baseline data as a guide to hypothesis formulation.

As a first step, ozone and radon data from 1989 were split into three groups by wind sector: The current baseline, south-west sector, a northerly sector receiving air from the Australian continent, about 400 km away, and a south-east sector receiving air from Tasmania. Because of seasonal variations, the data were further subdivided into monthly sets. Other factors known to affect ozone concentrations, are the presence of industrial pollution and wind speed (Galbally et al 1986). The effects of these will be considered separately.

Industrial pollution of air samples is reliably indicated by the presence of elevated CN concentrations. Thus the monthly data for each regional data set was split into two groups, "high CN" with CN higher than the CG baseline level of 600 cm^{-3} and "low CN" with CN below this level. A linear regression was performed between ozone and radon in each group, and the result expressed as "+" if there was a positive correlation between ozone and radon, "-" if negative, and "0" if there was no significant correlation. The results are shown in table 1.

Table 1: Summary of Regression analysis of Ozone versus Radon.

Sampling Conditions		Number of months in which the correlation was		
wind sector	CN	+	0	-
Baseline	Low	1	6	5
	High	5	3	4
Australian	Low	1	5	5
	High	5	3	4
Tasmanian	Low	0	2	10
	High	2	2	8

Some features of these results can be explained in terms of existing knowledge. Thus there is a striking difference between the Tasmanian sector and the others. The correlation is almost invariably negative, indicating that the predominant influence on ozone from this sector is local loss to the land surface. In winds from Australia, those with high CN are most likely to exhibit a positive correlation resulting from ozone input from industrial pollution exceeding the loss to the land surface.

One feature which was not anticipated is the strong similarity between the baseline and Australian sectors. It suggests that high radon events in baseline conditions may be more closely associated with intrusions of air from the Australian continent rather than Tasmania. Analysis of baseline records has shown that the highest radon levels usually occur within a few hours of a switch from non-baseline northerly wind conditions after passage of a front. The similarity of the ozone/radon relationship in the baseline and Australian wind sectors is therefore reasonable.

It is notable that CN concentration is very effective in selecting samples from the baseline wind sector where ozone and radon are positively correlated. This emphasises the importance of including CN as a baseline criterion.

As mentioned above, wind speed correlates with ozone concentration at inland sites. Galbally et al (1986) suggest that the mechanism for this is that wind speed is an indication of vertical mixing. At low wind speeds ozone destroyed at the land surface is not replaced fast enough to prevent a reduction in ozone concentration. A negative correlation could therefore be expected between ozone and local wind speed for winds from the Tasmanian sector. Indeed the ozone concentrations in samples with wind speed less than 30 km h^{-1} proved to be on average 2 ppbv lower than in samples with wind speed higher than 30 km h^{-1}.

Radon concentration is an indicator of contact with land, and table 1 shows that radon is generaly negatively correlated with ozone

in the Tasmanian sector. One way to compare wind speed to radon in selecting samples is to check the difference in ozone concentrations for high and low wind speeds in samples selected by radon concentration. The result is that wind speed has very little effect on ozone concentrations if the radon concentrations of samples is the same, the average difference being only 0.6 ppbv, a quarter of the difference when radon was not used. The reverse does not hold: radon has a strong negative correlation with ozone whatever the wind speed range selected. At CG it would therefore appear that radon is a better indicator of ozone loss than wind speed.

The answer to the original question about the effect of continental air on baseline ozone samples is that there are observable differences between ozone concentrations measured in samples from the baseline wind sector with elevated radon or CN. For samples with high radon in the absence of CN, the values are more frequently depressed than elevated. High CN indicates the possibility of ozone generation from industrial pollution. In this case the ozone may increase or decrease according to whether the destruction by contact with land exceeds the production.

6. Quasi-real time radon estimation

An alternative to measuring radon is to measure the radioactive decay products known as radon daughters. The main advantage is that it is feasible to build a sensitive instrument with a much shorter time response than is possible for radon. The reason for correlation of daughter to radon ratio to CN is that daughters are produced as small ions or ion clusters, some of which become attached to CN. When attached they are much less likely to be lost to the ocean than the small ions. Whittlestone (1990) has demonstrated that conditions in the marine boundary layer lead to quite high proportions of daughters being lost over to the ocean, especially when CN concentration is low. This means that daughter measurements are not useful for quantitative indication of land contact or for transport studies. However, at concentrations appropriate for a baseline threshold, there is a fairly close relationship between CN and daughter to radon concentration ratio. The present study investigates the possibility of using a combination of radon daughter and CN concentrations to derive an estimate of radon concentration which could serve as a baseline indicator.

Data were collected from July 16 to October 20 1991. For a quarter of this period all instruments were operational and the wind was in the baseline sector. The aims were: to see how effectively radon could be estimated from GADD results in combination with CN data; and to evaluate the performance of the estimation as a baseline criterion.

It was essential to evaluate the daughter measurements in terms of radon, the detection of which was relatively slow. All the data were therefore formed into hourly averages and the radon results brought forward 90 minutes to allow for the time lag in the detector response. The few occasions when the rate of change of the radon

concentration was too fast to permit realistic comparison of the daughter with the radon measurements were excluded.

An empirical models was derived from the measured concentrations of radon, its daughters and CN. Previous measurements at Cape Grim (Whittlestone 1990) have shown that an accuracy of about 30% can be achieved when CN are used to estimate the rate of loss of daughters to the ocean.

The logarithm of CN concentration was approximately linearly related to the daughter to radon ratio, and the resulting estimate of radon, RCN, using GADD counts, GD, with correction for the effect of CN was:

$$RCN = 38 + GD/(0.773 \times LOG(CN) - 0.0963) \quad \text{for CN} <= 300 \qquad 1a)$$
$$= 38 + GD/(3.13 \times LOG(CN) - 5.92) \quad \text{for CN} > 300 \qquad 1b)$$

where CN has units of number cm^{-3}

One indication of the success of estimating radon from GADD counts was to correlate radon with the estimates from equation 1. At 0.93, the correlation coefficient was considered good.

Since the radon estimate appeared to be satisfactory, it was appropriate to proceed to the next stage of checking how it would perform compared with radon as a baseline criterion. A radon threshold of 100 mBq m^{-3} was set and the GADD estimates divided into groups with radon higher and lower than the threshold. Above and below threshold events were selected correctly by GADD 85% and 95% of the time respectively.

7. Baseline selection

Having determined that GADD can provide rapid, accurate indication of whether the radon levels are above or below a threshold value, it was possible to assess the practical value of including such data in a baseline selection procedure. Figure 3 shows some time sequences of radon (from equation 1) and CN. Where there are periods of non-baseline wind direction, the data are left out, leaving a 4 hour gap. In the top graph, A, there are two periods immediately after a change from non-baseline when radon is higher than the 100 mBq m^{-3} baseline threshold while CN is below its 600 cm^{-3} threshold. The opposite happens in B, when the radon is low while CN is high.

These cases were typical of the four month period studied. Above threshold radon occurred in the baseline wind sector most often immediately after a period of non-baseline. These conditions are often the result of a front passing over the station accompanied by rain. This offers a possible reason for the low CN with the high radon. At the end of a baseline period, it is not uncommon for the wind to drift round to pass close to the coast, which is likely to yield CN without a strong radon signal, as seen in Figure 3B.

One consequence of the tendency of elevated radon in the baseline wind sector to occur after a non-baseline period is that there are

very few occasions when a result would be required from GADD in less than 20 minutes. The main purpose of GADD is to permit rejection of non-baseline data without undue loss of good data. During the period studied, CN were a satisfactory baseline indicator for transitions from baseline to non-baseline. GADD can therefore contribute most usefully in providing an indication that the air has become clean after a period of non-baseline. Its time response of 20 minutes therefore would lead to only a small loss of data at the beginning of baseline periods.

To assess the effect of using the radon threshold for selecting baseline, the data from the baseline wind sector have been grouped into above and below threshold groups for both radon and CN (table 2).

Table 2: Performance of CN and radon as baseline criteria

	Number of Events		Percentage of Total	
Radon (mBq m^{-3})	<=100	>100	<=100	>100
CN (cm^{-3})				
July-October				
<=600	859	121	86	12
> 600	6	10	1	1
July-August				
<=600	416	70	85	14
> 600	1	0	0	0
Sept.-October				
<=600	443	51	85	10
> 600	5	10	1	2

To illustrate the effect of the seasonal variation of CN, the data are presented in three groups: the full 4 months, the first two months and the last two months. The proportion of events with high radon is much the same in the winter as the spring (about 12%) , but the higher baseline CN level in spring resulted in a marked increase in above threshold events compared to winter, from 0.2 to 3%.

Over the 4 month period studied, about 13% of the data from the baseline wind sector at CG would have been rejected, most of it just following a transition to the non-baseline wind sector. Since this period is highly variable, and frequent occurrence of rain likely to reduce the effectiveness of CN as a baseline indicator, it is clear that deployment of GADD output as one of the baseline criteria for Cape Grim should result in samples less affected by recent contact with land. There were periods of high CN with below threshold radon. This emphasises that CN should continue to be used as a baseline criterion. However, the seasonal variation of CN results in a variable

threshold. It is therefore strongly recommended that the CN threshold be adjusted seasonally to achieve a constant sensitivity to incursions of polluted air.

8. Conclusions

The application of radon as a selection criterion for methane has demonstrated that the data from the MLO radon instrument provide provide valuable insights into causes of variability of methane concentration. In particular, there is a strong indication that methane selected from times at which the radon is below 100 mBq m^{-3} is much better matched to the output of the global models of Fung et al(1991) than the full record. It is therefore likely the radon baseline criterion is necessary for verification of models for any gas influenced by passage over land.

At Cape Grim, the baseline sector receives less recent continental air than MLO. Even so, more than 10% of samples contain radon at concentrations higher than 100 mBq m^{-3}. In such samples the ozone concentration in high radon samples is likely to be as much as 10 ppbv different from the below threshold radon group. A good guide as to whether the perturbation will be positive or negative is the CN concentration.

At both baseline stations, radon can be used as an objective baseline criterion, able to indicate air of recent continental origin in samples. The ability to provide faster measurements of radon would therefore improve the quality of samples taken by conditional samplers.

GADD is able to provide an accurate, rapid indication that ambient radon concentration has exceeded a baseline threshold of 100 mBq m^{-3}. Satisfactory accuracy is achieved by using an algorithm involving the current CN concentration to estimate radon from the raw count.

11. Acknowledgements

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Figure Captions

Figure 1. Map showing the location of Cape Grim and Mauna Loa.

Figure 2. Monthly averages of methane at MLO, selected by radon concentration range. Samples during upslope winds have been excluded.

Figure 3. Radon (estimated from GADD) and CN concentrations at Cape Grim under baseline wind conditions, October 1991.

CAPE
GRIM
MACQUARIE IS.

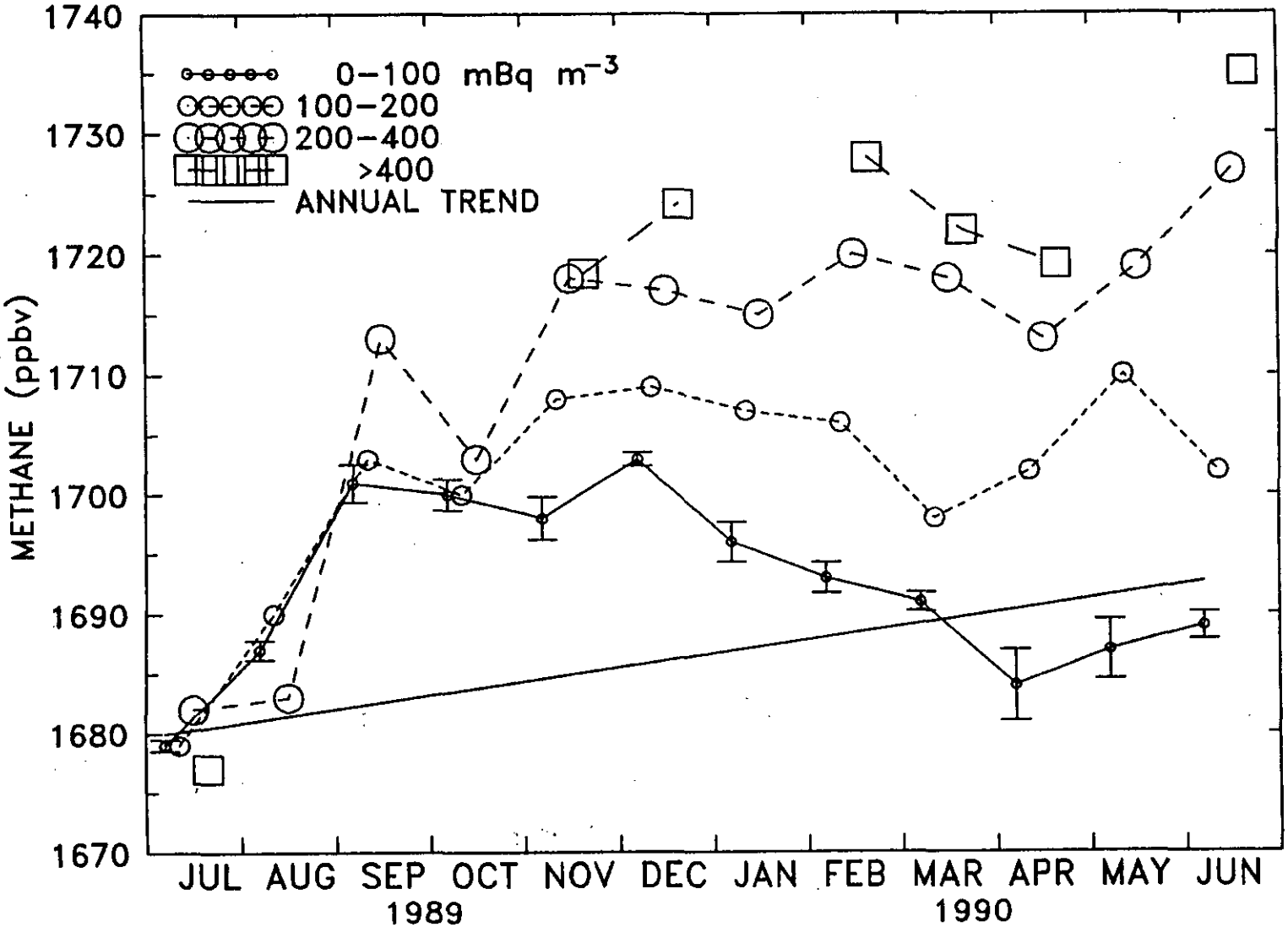

FIGURE 2. Monthly averages of methane at MLO, selected by radon concentration range. Samples during upslope winds have been excluded.

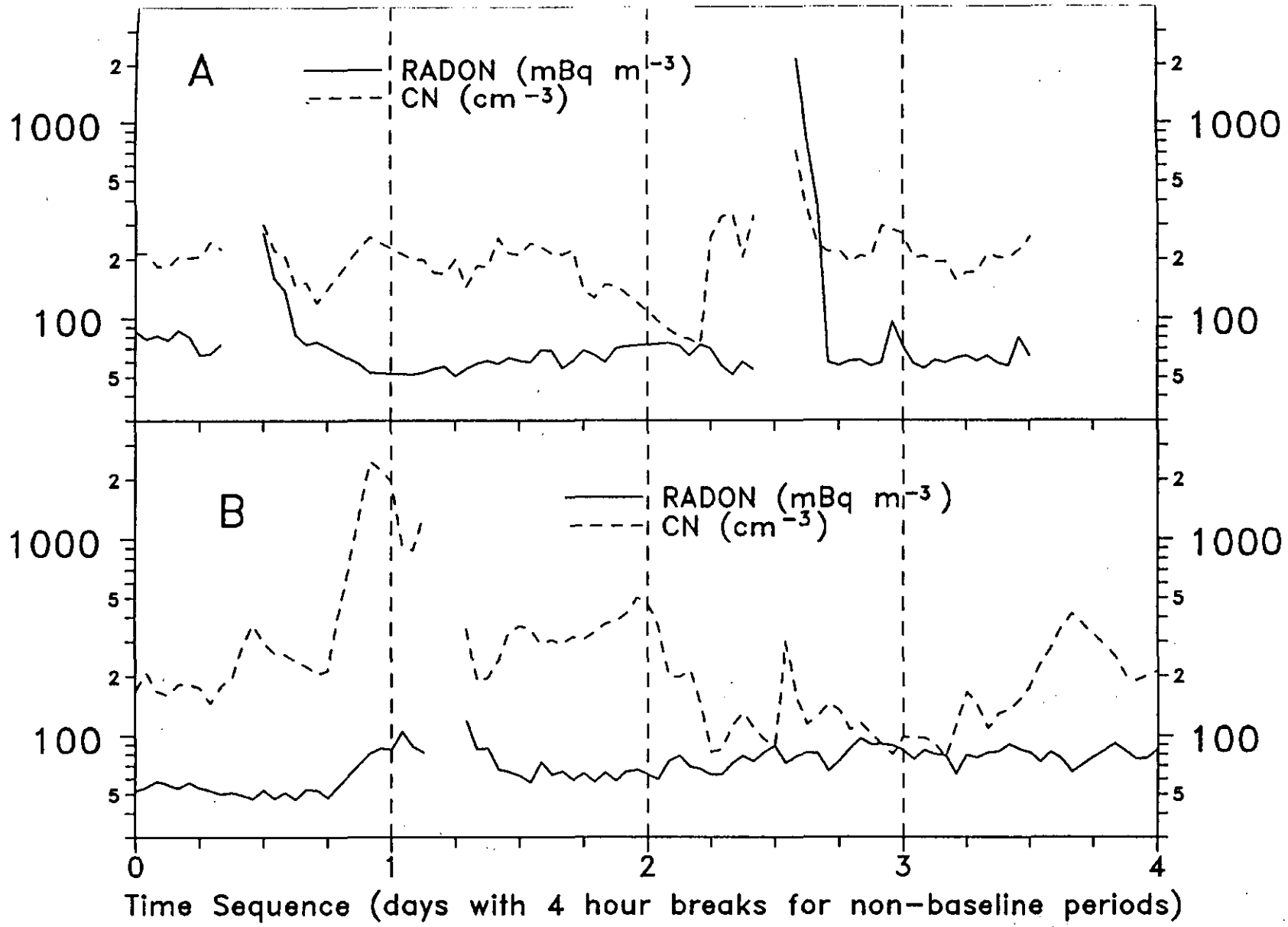

FIGURE 3: Radon (estimated from GADD) and CN concentrations at Cape Grim under baseline wind conditions, October 1991.