North American influence on atmospheric carbon dioxide data collected at Sable Island, Canada

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(Manuscript received 28 February 2002; in final form 2 December 2002)

ABSTRACT

Continuous and flask measurements of atmospheric CO₂ taken at Sable Island from August 1992 to April 1993 are presented and characterised as a function of air mass origin. The atmospheric environment over Sable Island (43°56'N, 60°01'W) is continuously influenced by the complex meteorology of synoptic systems moving off North America. This makes the interpretation of the Sable Island CO₂ data difficult. However, trajectory analysis shows distinct quantitative differences between the statistics of CO₂ measurements associated with air masses from "North America" (regions of high anthropogenic and terrestrial biospheric fluxes associated with much of the United States and the southern half of Canada) and of those associated with air masses from the "Arctic/North Atlantic" (regions of few terrestrial fluxes and oceans associated with the northern half of Canada and the Atlantic Ocean). When the continuous CO2 data are segregated into these two trajectory sectors for the period of observation, air masses originating in the North American sector show a higher CO₂ mixing ratio by ~2 ppm in winter and lower by \sim 3 ppm in summer, compared to air masses arriving from the other sector. Furthermore, the continuous Sable Island CO₂ measurements show a detectable monthly mean (August/September) diurnal cycle with an amplitude of ~2 ppm, with a minimum occurring on average around noon local time. Given the timing of the observed diurnal minimum and the lack of vegetation on the island, this indicates that the diurnal pattern observed at Sable Island is a diffused remnant of diurnal cycles transported from the main North American continent. These characteristic details are not captured by the discrete flask sampling program on the island.

1. Introduction

Recent studies based on atmospheric measurements from global networks indicate the presence of a large terrestrial sink for CO₂ on the order of 1–2 GtC yr⁻¹ in middle to high latitudes in the Northern Hemisphere (Tans et al., 1990; Enting and Mansbridge, 1991; Dixon et al., 1994; Conway et al., 1994; Denning et al., 1995; Ciais et al., 1995; Fan et al., 1998). Considerable effort is now focused on the estimation of magnitude and distribution of this terrestrial sink. One of the more common approaches to infer large-scale surface CO₂ sources and sinks utilizes inversions of the atmospheric CO₂ observations.

A relatively recent example of an inverse modeling study (Fan et al., 1998) used an integrated dataset for the period 1988 to 1992 (Masarie and Tans, 1995) and estimated a mean annual net North American uptake of CO_2 of 1.7 \pm 0.5 GtC yr⁻¹, which almost balances the annual emission of CO2 by fossil fuel burning in Canada and the United States. After eliminating the effect on atmospheric CO₂ distribution due to anthropogenic and oceanic processes, the estimate of the North American terrestrial sink of Fan et al. was based on a gradient in atmospheric CO₂ from the North Pacific to the North Atlantic of only -0.3 ppm (parts per million). Their study revealed that the inclusion or deletion of CO2 observations from Sable Island had a large impact on the final North American sink estimate. When the flask data from Sable Island obtained over the same 4-yr period (1988–1992) were included

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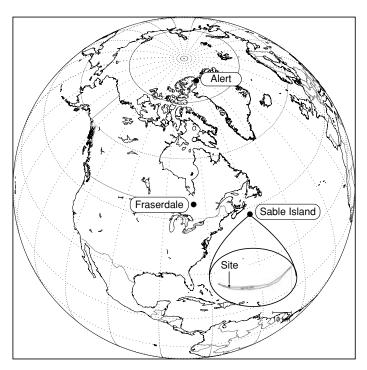


Fig. 1. Map showing locations of Sable Island (43°56'N, 60°01'W), Fraserdale (49°53'N, 81°34'W) and Alert (82°27'N, $62^{\circ}31'W$).

in their inverse calculation, the North American terrestrial sink estimate was reduced by 30%.

Given the obvious importance of Sable Island in terms of its location and data quality in a carbon budgetary investigation for North America, it is essential that we obtain insight into the nature of CO₂ measurements at Sable Island. In this paper we characterize and interpret a 9-month (August 1992 to April 1993) record of continuous and flask CO2 measurements taken at Sable Island by the Meteorological Service of Canada. The measurements show that CO₂ is highly variable and significantly influenced by air masses from the North American continent. One-day back-trajectories are used to evaluate the air mass origins and the effect of North America on the CO2 data at Sable Island. The relatively short in-situ NDIR CO2 record shows unique features that are not captured by the CO2 flask record, thus providing a valuable additional constraint for atmospheric carbon cycle models.

2. A description of the sampling location

Sable Island is a remote crescent-shaped "sandbar" located at 43°56′N and 60°01′W in the Atlantic Ocean, approximately 275 km east-southeast of Halifax, Nova Scotia (Fig. 1). The island is about 40 km long with little over 1 km wide at its widest point, and has maximum elevation of about 10 m. The terrain along the shoreline is flat, with the interior containing two large saltwater ponds and a series of rolling sand dunes covered with small patches of grass and shrubs. Thus, vegetation on the island is very sparse. Public access is restricted. There are 6-8 permanent residents on the island who are responsible for maintaining the Environment Canada weather and science observational programs. The island is also a refuge for wild horses, seals and migratory birds. The only major local source of pollution is from the diesel generators located approximately 180 m east of the weather station that is located approximately 8 km from the western end of the Island, $100\,\mathrm{m}$ from the north beach and $800\,\mathrm{m}$ from the south beach.

Sable Island is predominantly in the flow of air coming off the North American continent. However, because it is located in an area where offshore continental air masses mixes with warm tropical air mass influenced by the Gulf Stream, synoptic weather events and the boundary layer dynamics over Sable Island are

complex and air mass trajectory analysis of CO2 measurements taken on the island can be problematic. Yet, as will be shown later in the paper, there are signals in the statistical character of CO2 to indicate that the Sable Island measurements do in fact "see" at least some aspects of the continental CO2 behaviour on diurnal to monthly time scales.

3. Continuous CO₂ measurements

In August 1992, a pilot study was initiated at Sable Island to evaluate the large variability observed in the flask CO₂ measurements taken on the island since 1975 (Wong et al., 1984; Chung, 1988). A continuous NDIR (URAS-3E, Hartman & Braun) measurement system was installed in the attic of the upper air station building, and was in operation until April 1993. Ambient air was pumped through a 7- μ m stainless steel membrane filter to the NDIR at approximately 5 L min⁻¹ using a 0.95-cm OD sample line (Dekoron) that extended to the top of a 15-m tower located about 0.5 km from the southern shoreline. The sample air was dried to a dew point of around -60 °C by first passing it through a glass coil refrigerated at 0 °C (to remove most of the excess water) and then through a glass trap submerged in a -80 °C methanol bath.

A linear response function of the analyzer was determined every hour by passing two calibrated CO₂in-air standard gases through the analyzer on the hour for 5 min each. The two CO₂ standards were chosen to bracket the range of the expected ambient CO₂ mixing ratios. Ambient air was passed through the system for the remaining 50 min. Outputs from the analyzer were averaged over 5-min periods. Ambient CO₂ values were determined using the calculated linear response function of the analyzer. The two bracketing air standards were calibrated at the site against a set of five CO₂-in-air field standards every 8 d. These field standards were calibrated in Toronto before being deployed to the station. All the CO_2 measurements used in this study have been adjusted to the WMO mole fraction scale established and maintained at the Central Calibration Laboratory at NOAA in Boulder. Colorado (Zhao et al., 1997). All CO₂ measurements are reported in μ mol mol⁻¹ (abbreviated ppm mixing ratio) dry air.

Hourly-averaged atmospheric CO₂ measurements from Sable Island for the period 1500 GMT, 3 August 1992 to 1800 GMT, 4 April 1993 are shown in Fig. 2. The CO₂ data from the flask sampling program are also displayed. [A brief description of the flask sam-

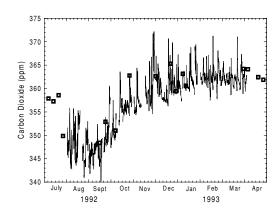


Fig. 2. Hourly averaged CO₂ mixing ratios (ppm) for the period 1500 GMT, 3 August 1992 to 1800 GMT, 4 April 1993. The data have been edited to exclude CO₂ measurements that may be affected by contamination from local pollution souces and when the analytical system was not functioning within operational limits. Also plotted are flask CO₂ mixing ratios (□) sampled during the same period.

pling protocol at Sable Island is given later in the paper. A detailed description of the flask sampling program at Sable Island is given in the Meteorological Service of Canada (MSC) report Canadian Baseline Program: Summary of Progress to 1998 (MSC, 1999).] It is clear that the low resolution of the CO₂ flask record (typically less than one sample per week) limits the information available for analysis of short-term variations and identification of periods of long-range transport. The rest of the paper will focus on the statistical characterization of the results from a trajectory analysis of the continuous data shown in Fig. 2. (We will, however, briefly discuss flask data later in Section 4.) The data set does not include values when local wind direction indicated the possibility of local anthropogenic contamination. It should be noted that the following discussion and results pertain only to the period of insitu CO₂ measurements, and any extrapolation to the period outside of the relevant time domain is unwarranted.

3.1. Continental influence: seasonal cycle

The location of Sable Island provides an excellent platform for observing atmospheric transport of anthropogenic pollutants such as carbon monoxide and ozone from North America (Parrish et al., 1993; 1998), and is ideal for assessing the influence of continental plumes in marine boundary-layer processes (Fehsenfeld et al., 1996; Duderstadt et al., 1996; Wang et al., 1996). These studies indicate clearly that the

atmospheric composition over Sable Island is influenced significantly by the transport of emissions from North America.

The seasonal and secular changes in the CO₂ flask sampling record at Sable Island have been published previously (Wong et al., 1984; Nakazawa et al., 1997), but only one study has specifically attempted to link the observed variability in the CO₂ data record at Sable Island with respect to air mass origin (Chung, 1988). That study used air parcel trajectory analysis to show that many of the "low" CO2 values in 1984 were associated with air parcels coming generally from the north or east-southeast. In contrast, relatively "high" values were found to be associated with trajectories from the west or west-southwest, i.e., from the North American continent.

The influence of continental CO₂ fluxes on the Sable Island data is evaluated by performing air mass trajectory analysis on the 9 months of continuous CO₂ data shown in Fig. 2. To facilitate the analysis procedure, trajectories are classified into two sectors, as shown in Fig. 3. Sector 1 contains the major North American anthropogenic and terrestrial source/sink areas, and the trajectories originating in this sector will be designated as "North American." Air parcels originating in Sector 2 will be called "Arctic/North Atlantic." A 6-h mean CO₂ mixing ratio is calculated from the hourly averaged CO2 mixing ratios at Sable Island. Endpoints of

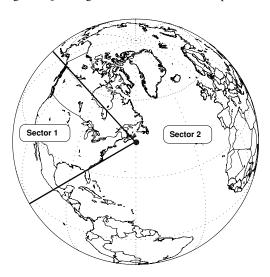


Fig. 3. Sable Island trajectory sectors associated with different source regions. The trajectory endpoint sector classification scheme uses a transformed coordinate system with Sable Island defined as the centre. Sectors 1 and 2 are defined as the "continental" and "non-continental" sectors, respectively.

the averaging periods coincide with the arrival times of the air mass represented by the trajectory calculations. These mean CO2 values are then assigned to one of the two sectors.

Using the hemispheric MSC trajectory model (Olsen et al., 1978), 1-d Lagrangian back trajectories are calculated every 6 h at 0000, 0600, 1200 and 1800 UTC for air parcels reaching Sable Island at the endpoint pressure level of 925 hPa. The trajectory model is three-dimensional, and uses wind and temperature data archived on standard pressure levels at the Canadian Meteorological Centre (CMC). The 925 hPa trajectories are applied in this study because they are considered to be most representative of the origin of air sampled at Sable Island. In general, Sable Island trajectories calculated for the pressure levels of 925 and 850 hPa are quite similar.

Six-hourly averaged CO2 values segregated by sectors are shown in Fig. 4. As already mentioned above, Sector 1 is assumed to represent significant North American influence, showing regional anthropogenic and biological impact on the CO₂ measurements at Sable Island. The time series of the two sectors are fitted with fifth-order polynomial curves, which minimize the root mean square (RMS) errors. By applying Student's-t-test to the monthly mean values, the two time series are found to be significantly different at the 5% level. By applying the F-test it is also found that the variability of CO₂ in Sector 1 is significantly greater than that in Sector 2, although the variability becomes less and similar in magnitude in March for both sectors. It is also interesting to note that, at least for the period of observation, the CO₂ variability in August to October period is greater than in January to March, again statistically significant at the 5% level. This is in contradiction with the result that no seasonal dependence with regard to the observed CO₂ variability was found in the flask data (Chung, 1988).

The most striking feature in the two CO₂ time series plots in Fig. 4 is that during August and early September, CO₂ mixing ratios in air parcels arriving from Sector 1 are significantly lower (by \sim 3 ppm) than those associated with air parcels from Sector 2, suggesting that the terrestrial uptake of CO₂ due to photosynthetic activity is detectable at Sable Island due to its proximity to the seasonal CO₂ source/sink regions in North America. With the termination of photosynthesis in the late fall, the impact of North American biospheric respiration on atmospheric CO₂ is evident at Sable Island during late November to early December, with Sector 1 showing a CO₂ mixing ratio about 3 ppm higher than

Fig. 4. Six-hourly averaged CO2 data for the study period. The data are segregated into two time series of CO2 values, one associated with Sector 1 trajectories (●) and the other with Sector 2 trajectories (□). Sector 1 (continental) and Sector 2 (non-continental) time series are fitted with fifth-order polynomial curves represented by thick and thin lines, respectively.

that associated with Sector 2. The minimum associated with Sector 2 appears to be more representative of an atmospheric CO2 field in a marine environment, the so-called "background" concentration field.

Figure 5 compares the curves for Sable Island (Fig. 4) with similar curves for Alert and Fraserdale. Fraserdale (49°53′ N, 81°34′W) is located in a north-

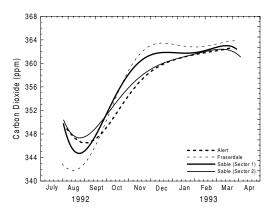


Fig. 5. Two CO₂ polynomial curves from Fig. 4 are re-plotted and compared to curves from Alert and Fraserdale (see Fig. 1 for geographical location) generated by a fifth-order polynomial fit to Alert (1988-1998) and Fraserdale (1990-1996) data, respectively.

ern Ontario region characterized by extensive wetland and boreal forest ecosystems. A detailed description of the Fraserdale measurement site can be found in MSC (1999). Alert (82°27′N, 62°31′W) is located on the northeastern tip of Ellesmere Island in the Canadian Arctic. The smoothed curves for Alert and Fraserdale shown in Fig. 5 are generated using the fifth-order polynomial fit through the continuous CO₂ data obtained at Alert (1988-1998) and Fraserdale (1990-1996). Due to CO₂ diurnal variability at Fraserdale, only the afternoon data, when the convective boundary layer is likely to be well mixed, are used in this analysis. No diurnal signals are observed in the Alert data.

A qualitative inspection of Fig. 5 shows that the seasonal cycles at Fraseradale and Sable Island (Sector 1, North America) show a greater degree of similarity than those at Alert and Sable Island (Sector 2, Arctic/North Atlantic). Although Fraserdale shows a greater seasonal amplitude, it is similar in phase to Sector 1 at Sable Island. The Fraserdale seasonal minimum in late August is about 3 ppm lower than that for Sector 1, owing to the strong biogenic influence in summer at the site. The rates of the atmospheric CO₂ rise observed during the autumn at Fraserdale and in Sector 1 at Sable Island are similar, and reflect the relatively fast termination of photosynthesis on the

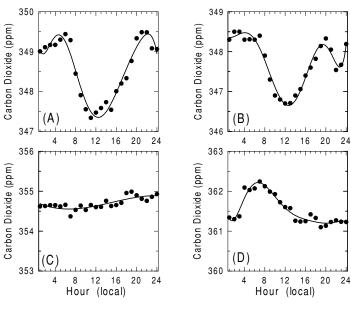


Fig. 6. Monthly averaged diurnal cycles for (A) August, (B) September, (C) October and (D) January.

continent, going from net CO_2 uptake to net release. This similarity between Fraserdale and Sector 1 at Sable Island is consistent with the existing hypothesis of atmospheric transport of the terrestrial seasonal signal to Sable Island from the continent. The seasonal evolution of CO_2 for Sector 2 shows a greater degree of similarity to that observed at Alert than to those associated with Fraserdale and Sector 1. This similarity in the seasonal cycle shown in Fig. 5 between Alert and Sector 2 is consistent with an earlier statement regarding Sector 2 representation of the background atmospheric CO_2 concentration field.

3.2. Continental influence: diurnal cycle

Diurnal cycles in CO₂ are consistently observed during the growing season at continental sites such as Fraserdale (MSC, 1999). In summer, CO₂ mixing ratios have maximum values in the morning (typically around 06:00 to 08:00 local time) and then decrease rapidly to reach minimum values in the late afternoon to early evening (typically around 16:00–19:00 local time). The diurnal amplitude is typically 30–50 ppm, sometimes reaching as high as 80 ppm, at the 40-m level on the tower. Another good example of a description of a daily CO₂ evolution at a continental site can be found in Bakwin et al. (1995), who describe

continuous CO_2 measurements from four levels on a 500-m tower in North Carolina, US.

Monthly mean diurnal cycles of the Sector 1 CO₂ at Sable Island are shown in Fig. 6 for August-October 1992 and January 1993. Clearly, the amplitude of diurnal cycle is strongly seasonally dependent. Whereas the diurnal cycle is nearly absent in October, it is quite evident in August and September, with a minimum occurring in the late morning to early afternoon and with an amplitude of \sim 2 ppm. Thus, when compared to observed diurnal cycles at continental sites that are influenced by local and regional vegetation, like Fraserdale, the diurnal amplitude at Sable Island is an order of magnitude smaller, and the timing of the cycle at Sable Island is such that the diurnal minimum occurs around noon local time. Since Sable Island is sparsely vegetated, the only logical conclusion as to the origin of the diurnal cycle observed on the island is the biospheric activity on the continent; it is the remnant of the diurnal cycle from the continent modified by advection. In order to validate this statement, we perform a simple idealized numerical experiment using a regional dynamical transport model called Mesoscale Compressible Community model (MC2) (Benoit et al., 1997). It is a dynamical meteorological model with comprehensive state-of-the-art physics package. For our study here, the model is integrated with a grid resolution of 20 km and in hydrostatic mode. The model is Tellus.cls

initialised and forced at the boundary with archived Canadian Meteorological Centre (CMC) meteorological analyses.

With MC2, we perform a case study of 6-8 August 1992, during which time the winds shift from continental to oceanic. The sole purpose of this numerical experiment is to demonstrate, with a prescribed biospheric function on the continent, that the variability in the CO₂ diurnal cycle at Sable Island is possible by atmospheric transport alone. It is not performed in order to simulate the results shown in Fig. 6. The domain of the model covers a North American region defined by latitudes 26.7°N to 77.2°N and by longitudes 235.4°E to 304.6°E. The diurnal CO₂ flux is assigned over the North American continental region at the bottom of the mesoscale model. It is defined for each land point in the model to have photosynthetic uptake between local sunrise and sunset with a functional form of the positive part of the sine function, with a total daily uptake of 7 g C m⁻². The soil respiration has a daily total of 3.4 g C m⁻², with a 20% sinusoidal diurnal variation about the mean value $(0.0394 \text{ mg C m}^{-2} \text{ s}^{-1})$. The maximum soil respiration is at 3 pm local time. There is no CO₂ flux from the ocean region. The initial atmospheric concentration of CO2 in the model is assigned an arbitrary value of 0 ppm. Given the westerly flow on 6 August the CO₂ signal from the continent reaches Sable Island after one day. Figure 7 shows the diurnal cycles in CO₂ concentration at 15m level at Sable Island for 7 and 8 August, as well as the average of these two days. Although the flow on 7 August is westerly, the long transport from the continent has highly modified the diurnal cycle. The magnitude is reduced to about 3.5 ppm, with a primary maximum at 3 am and a secondary maximum at 3 pm local time. The diurnal minimum occurs around 8-9 am. The flow on

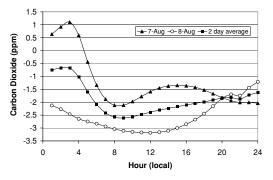


Fig. 7. Diurnal cycles of CO₂ concentration at 15m level simulated for 7 and 8 August, along with the average of these two days.

8 August is south-westerly, thus forcing the air from the continent to travel longer over the ocean before reaching Sable Island. The difference in the flow direction has resulted in a very different diurnal cycle for 8 August with much reduced amplitude (1.8 ppm) and a noticeable change in the timing of the occurrences of maximum and minimum. The 2-d average shows that the averaging process greatly smoothes the variability in the diurnal CO₂ variations. This is consistent with the smoothing of the observed large variability in the CO₂ concentration data that results in relatively small monthly averaged diurnal cycles shown in Fig. 6. An examination of the day-to-day change in the observed diurnal cycle at Sable Island in August and September shows a large variability in amplitude and phase, depending on the wind direction, and the timing of the occurrences of maximum and minimum shown in Fig. 7 is within that range of observed variability. The model results for this particular case demonstrate clearly that the diurnal cycle observed at Sable Island is almost certainly a remnant of the continental biospheric cycle.

An early morning 1-ppm CO₂ rise which appears to occur on many days in January (Fig. 6d) is not statistically significant at the 5% level. However, a possible anthropogenic CO₂ contribution from the continent to this phenomenon cannot be ruled out. Many of the detailed aspects of the diurnal cycle observed at Sable Island are now under investigation.

4. Flask samples

At this point in the presentation, we believe it is of interest to briefly discuss the scientific value of CO2 flask samples taken at Sable Island, since this program has been in effect since 1975 as part of the then WMO Background Air Pollution Monitoring Network (BAPMoN). We do this within a comparative context between flask and in-situ values shown in Fig. 2. However, before we proceed with the comparative discussion, we give a short description of the flask sampling protocol that has been employed at Sable Island, and in fact at all Canadian background stations.

In 1975, measurements of atmospheric CO₂ collected approximately once per week in discrete Pyrex glass flasks were established at Sable Island. The CO₂ mixing ratio in the flasks was initially analyzed at the Institute of Ocean Sciences (IOS) in Patricia Bay, British Columbia (Wong et al., 1984). In 1988, MSC in Toronto assumed responsibility for the CO₂ mixing ratio analysis. The flasks are 2 L in volume and are fitted with 6-mm bore high vacuum stopcocks lubricated with Apiezon-N grease. The flasks are first evacuated to a pressure of approximately 10⁻⁴ mbar at the main laboratory using a large vacuum pump before being deployed to the station. Flasks are sampled by having the operator simply opening the stopcock of the flask while walking into the wind. The stopcock is closed approximately 5-10 s later. A second additional flask is immediately sampled using the same procedure. The sampling time and applicable meteorological information are recorded and included with the shipment of flasks to MSC for analysis.

Prior to 1986, the air samples were always collected upwind of the station. From 1986, the sampling protocol was changed, and the air samples have been collected on the windward shore of the island. This has prevented the sampling of air that may have been influenced by local contamination or by the vegetation on the island. Flask samples are usually collected between 13:00 and 16:00 local time. The air samples are analyzed at MSC using NDIR methodology. A detailed description of the sampling program and analysis procedures is summarized in MSC (1999) and in Higuchi et al. (1987).

During the period of in-situ CO₂ measurement program, the accepted number of flask samples totaled 11. Had the sampling protocol been followed during this period at Sable, there would have been about 30 flask samples. However, only 15 samples were taken, and out of these, 11 were categorized as A (i.e., free of obvious contaminants, sampling and laboratory analysis errors). When compared with the closest hourly averaged in-situ data, these flask values show an average difference of 0.63 ± 0.66 ppm, with nine flask values being greater than the corresponding hourly in-situ values and two flask values being less. Three of the 11 flask values are more than 1 ppm higher than the corresponding hourly in-situ values. The observed offset is comparable to the offset observed by NOAA/CMDL (Conway et al., 1994). For Sable Island, the discrepancy between the flask and continuous data results partly from improper field flask sampling procedures. It was also found that the evacuated, 2-L greased stopcock flasks (GSF) used at Sable during this period proved to be problematic for field sampling. In a comparison study carried out between GSF and in-situ measurements at Alert, values from GSF showed a significant positive bias (0.91 \pm 0.72 ppm) since mid-1991 (Hudec and Trivett, 1995).

The primary objective of any flask sampling program is to capture secular trend, as well as inter-annual

variations in the seasonal cycle. In some cases, and in particular at Sable Island, the latter objective can be problematic due to inadequate sampling frequency. Even a cursory examination of Fig. 2 shows that a curve fit to the flask data at Sable Island from August 1992 to April 1993 would lead to some bias in the seasonal variation over this period. Flask samples during the period of minimum CO₂ values in August 1992 are missing, as well as during the period from the latter half of January to the first half of April 1993. Since inter-annual variations in the seasonal cycle is sensitive to the seasonal minimum in mid to high latitudes, it is essential that this feature is adequately captured by the flask sampling program if its data are to be used by carbon cycle models to estimate regional carbon sources and sinks. Since Fig. 2 is reflective of a typical sampling at Sable Island, it brings into question the usefulness of the flask data taken there. The results of our study and of Fan et al. (1998) have led us to implement a continuous CO₂ monitoring program at Sable Island, to ensure adequate sampling to resolve the features in atmospheric CO₂ that are reflective of biospheric activities in North America.

It is these details, however, that are precisely what is needed by any modelling effort to calculate terrestrial biospheric sources and sinks. In the case of Sable Island, for example, Fan et al. (1998) were able to reduce their North American biospheric sink estimate of $1.7 \,\mathrm{GtC}\,\mathrm{yr}^{-1}$ by 30% by including the interpolated data from a curve fit to the Sable Island flask data in their inverse modelling calculation. We highly speculate that the 30% change is due primarily to the flask sampling frequency being inadequate to capture a representative signal, particularly in summer. It is interesting to speculate how much the result of Fan et al. (1998) would have changed if a more representative CO₂ data set from Sable Island had been available.

5. Conclusions

Continuous CO₂ measurements from Sable Island were presented and discussed within the context of North American continental influence. Using trajectory analysis, we have shown that the CO₂ mixing ratios observed on Sable Island reflect temporal behaviour of CO₂ sources and sinks in North America.

The continuous CO₂ data were divided into "North American" and "Arctic/North Atlantic" sectors by the trajectory analysis. The CO₂ associated with air coming from the North American sector was found to be higher at Sable Island by about 2 ppm in winter

and lower by about 3 ppm in summer, when compared to CO₂ values associated with air from the Arctic/North Atlantic sector. In the North American sector data, a diurnal cycle with amplitude of about 2 ppm was detected during summer, with minimum occurring about 20 h later than those observed at continental sites in North America. Given the timing of the observed diurnal minimum and the lack of vegetation on the island, this indicates that the diurnal pattern observed at Sable Island is a diffused remnant of diurnal cycles transported from the continent upstream. That this is in fact the case was demonstrated by a numerical simulation of an atmospheric dynamical transport model with prescribed diurnal CO₂ flux. These seasonal and diurnal characteristics were not captured by the discrete flask sampling program on the island.

6. Acknowledgments

We thank Senen Racki for programming assistance and Tatiana Motchalova for the data sectoring analysis. We also thank Gerry Forbes and his staff of the weather station on Sable Island thanks for their conscientious effort in ensuring that good flask samples were obtained. Neil Trivett is gratefully acknowledged for his leadership in the greenhouse gas measurement program during his association with the Meteorological Service of Canada. We are also indebted to Ed Dlugokencky and Fred Hopper, who reviewed an earlier version of the paper; their comments and suggestions greatly improved the quality and readability of the original draft of the paper. Comments and suggestions by two anonymous reviewers are also much appreciated.

REFERENCES

- Bakwin, P. S., Tans, P. P., Ussler, W. III and Quesnell, E. 1995. Measurements of carbon dioxide on a very tall tower. Tellus 47B, 535-549.
- Benoit, R., Desgange, M., Pellerin, P., Pellerin, S., Desjardins, S. and Chartier, Y. 1997. The Canadian MC2: a semi-Lagrangian, semi-implicit wide-band atmospheric model suited for fine-scale process studies and simulation. Mon. Wea. Rev. 125, 2382-2415.
- Chung, Y. S. 1988. The variations of atmospheric carbon dioxide at Alert and Sable Island, Canada. Atmos. Environ. 22 383-394
- Ciais, P., Tans, P. P., Trolier, M., White, J. W. C. and Francey, R. J. 1995. A large northern hemispheric terrestrial CO₂ sink indicated by the $^{13}\text{C}/^{12}\text{C}$ ratio of atmospheric CO₂. Science 269, 1098-1102.
- Conway, T. J., Tans, P. P., Waterman, L. S., Thoning, K. W., Kitzis, D. R., Masarie, K. A. and Zhang, N. 1994. Evidence for interannual variability of the carbon cycle from the NOAA/CMDL global air sampling network. J. Geophys. Res. 99, 22831-22855.
- Dixon, R. K., Brown, S., Houghton, R. A., Soloman, A. M., Trexler, M. C. and Wisniewski, J. 1994. Carbon pools and flux of global forest ecosystems. Science 263, 185-
- Denning, A. S., Fung, I. Y. and Randall, D. 1995. Latitudinal gradient of atmospheric CO2 due to seasonal exchange with land biota. Nature 376, 240-243.
- Duderstadt, K. A., Carroll, M. A., Sillman, S., Wang, T., Albercook, G. M., Feng, L., Parrish, D. D., Holloway, J. S., Fehsenfeld, F. C., Blake, D. R. and Forbes, G. 1996. Photochemical production and loss rates of ozone at Sable Island, Nova Scotia during the North Atlantic Regional Experiment 1993 summer intensive. J. Geophys. Res. 103,
- Enting, I. G. and Mansbridge, J. V. 1991. Latitudinal distri-

- bution of sources and sinks of CO2 results of an inversion study. Tellus 43B, 156-170.
- Fan, S.-M., Gloor, M., Mahlman, J., Pacala, S., Sarmiento, J., Takahashi, T. and Tans. P. 1998. A large terrestrial carbon sink in North America implied by atmospheric and oceanic carbon dioxide data and models. Science 282, 442–446.
- Fehsenfeld, F. C., Daum, P., Leaitch, W. R., Trainer, M., Parrish, D. D. and Hubler, G. 1996. Transport and processing of O₃ and O₃ precursors over the North Atlantic: An overview of the 1993 North Atlantic Regional Experiment (NARE) summer intensive. J. Geophys. Res. 101, 28877-28891.
- Hudec, V. and Trivett, N. B. A. 1995. An evaluation of CO₂ flask measurement programs at Alert, N.W.T. In: Report of the 8th WMO Meeting of Experts on Carbon Dioxide Concentration and Isotopic Measurement Techniques (ed. T. Conway). WMO Global Atmosphere Watch, No. 121, 42-57.
- Higuchi, K., Trivett, N. B. A. and Daggupaty, S. M. 1987. A preliminary climatology of trajectories related to atmospheric CO₂ measurements at Alert and Mound Bay. Atmos. Env. 21, 1915-1926.
- Masari, K. A. and Tan, P. P. 1995. Extension and integration of atmospheric carbon dioxide data into a globally consistent measurement record. J. Geophys. Res. 100, 11593-11610.
- MSC, 1999. Canadian Baseline Program: Summary of Progress to 1998. Air Quality Research Branch Report, Meteorological Service of Canada, Toronto, Ontario,
- Parrish, D. D., Holloway, J. S., Trainer, M., Murphy, P. C., Forbes, G. L. and Fehsenfeld, F. C. 1993. Export of North American ozone pollution to the North Atlantic Ocean. Science 259, 1435-1439.
- Parrish, D. D., Trainer, M., Holloway, J. S., Yee, J. E., Warhawsky, M. S., Fehsenfeld, F. C., Forbes, G. L. and

- Moody, J. L. 1998. Relationships between ozone and carbon monoxide at surface sites in the North Atlantic region. *J. Geophys. Res.* **103**, 13 357–13 376.
- Nakazawa, T., Ishizawa, M., Higuchi, K., Trivett, N. B. A. 1997. Two curve fitting methods applied to CO₂ flask data. *Environmetrics* **8**, 197–218.
- Olsen, M. P., Oikawa, K. K. and MacAfee, A. W. 1978. A trajectory model applied to the long-range transport of air pollutants: a technical description and some model inter comparisons. *Environment Canada/AES Internal Report* LTRAP 78-4, Toronto, Ontario, 24 pp.
- Tans, P. P., Fung, I. Y. and Takahashi, T. 1990. Observational constraints on the global atmospheric CO₂ budget. *Science* **247**, 1431–1438.
- Wang, T., Carroll, M. A., Albercook, G. M., Owens, K. R., Duderstady, K. A., Markevitch, A. N., Parrish, D. D., Holloway, J. S., Fehsenfeld, F. C., Forbes, G. and Ogren, J. 1996. Ground-based measurements of NO_x and total reactive oxidized nitrogen (NO_y) at Sable Island, Nova Scotia, during the NARE 1993 summer intensive. *J. Geophys. Res.* 101, 28 991–29 004.
- Wong, C. S., Chan, Y.-H., Page, J. S., Bellegay, R. S. and Petit, K. 1984. Trends of atmospheric CO₂ over Canadian WMO background stations at Ocean Weather Station P, Sable Island and Alert. *J. Geophys. Res.* **89**, 9527–9539.
- Zhao, C. L., Tans, P. P. and Thoning, K. 1997. A high precision manometric system for absolute calibrations of CO₂ in dry air. J. Geophys. Res. 102, 5885–5894.