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Tropospheric transport of trace substances in the southern hemisphere

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ABSTRACT

The mean annual cycle of elemental carbon (EC) has been deduced from a 7-year series of measurements made at Cape Grim, Tasmania. The most conspicuous features are a spring (October) maximum and summer (January) minimum. These are also found in the mean annual cycle at Cape Grim of carbon monoxide thought to be produced at the same time and in the same regions (predominantly much further north). Differences in detail in the cycles are consistent with the different removal mechanisms involved. However, the very considerable similarities between the two curves suggest that seasonal variations on OH radical concentration is not the main cause of CO's seasonal cycle and also that seasonal variations in EC scavenging is not the main cause of EC's seasonal cycle. The seasonal variation of radon, also of continental origin, is out of phase with the variations of EC and CO, showing that strength of poleward transport is not the cause of the seasonal change of EC and CO. Methane, only a small portion of which is produced by biomass burning, has a seasonal variability closely similar to that of both EC and CO suggesting that biomass burning is the only source with significant seasonality of EC, CO, and CH_4. The broad summer maximum of mean ^7Be concentrations shows that the spring maximum of EC and CO is not due to upper transport with enhanced spring mixing. It seems unlikely that interhemispheric transport plays a major role in the seasonal cycle of EC, CO and CH_4 because of the phase lag between the predominantly northern hemisphere produced tracers and that of the three first named tracers. Comparison of the tracer cycles at Cape Grim, Cape Point, S. Africa and over Antarctica suggests that the spring maximum could be due to a peak in production of combustion products from biomass burning at low latitudes which is propagating throughout the southern hemisphere.

1. Introduction

Carbon monoxide, (CO), monitored at the Australian Baseline Atmospheric Pollution Monitoring Station at Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E) and at other mid- and high-latitude southern hemisphere sites has been shown by Fraser et al. (1986) to have interesting and consistent seasonal cycles. Fraser et al. (1986) attempted to model these changes from a knowledge of the likely sources and including variable transport and seasonal concentrations of the hydroxyl radical OH which limits the lifetime of CO in the atmosphere to only a few months. While the features of the annual cycle could be successfully reproduced in the models, knowledge of the actual variabilty of both transport and OH is very limited.

Comparison of the annual cycle of concentrations with that of any tracer produced by the same sources, having a lifetime in the atmosphere that is not too different and that is unaffected by the presence of OH would give us an opportunity to study the variability of OH through differences from OH-affected tracers. The fine particle portion of elemental carbon, (EC), is such a tracer. Both CO and EC are produced mainly by biomass burning and other combustion sources,

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(e.g., Crutzen et al., 1985), though the ratio of outputs will vary with the particular sources.

We have therefore deduced the annual cycle of EC from seven years of measurements (April 1982–January 1990) made at Cape Grim and compared it with the monthly average CO-concentrations given for that site by Fraser et al. (1988). Comparisons with the seasonal variations of other species at Cape Grim and other southern hemispheric sites have then allowed further deductions concerning the physical processes that determine the concentrations of the various trace substances.

2. The annual cycle of elemental carbon at Cape Grim

The measurements described by Heintzenberg (1985) consisted of the collection of particles smaller than about 2 μm radius from 600–1000 m^3 of air on latex fiber filters, leaching away the water-soluble material, then photometrically estimating the elemental carbon (soot) present. Sampling was carried out only when the stations "baseline switch" was operating, the criteria for which were (1) wind direction between 190° and 280°, (2) condensation nucleus concentration <600 cm^{-3}. Air from Tasmania or direct flow from Australia was excluded by these criteria.

A sample sometimes took two weeks to collect and sometimes two months and a particular sample was likely to include contributions from part or all of many days. To simplify the analysis, we assumed that sampling took place uniformly over the whole length of the collection period. The year was divided into 25 equal segments to correspond roughly with the minimum sampling period. The concentration C_i of EC in sample i was weighted by the factor n_i in each segment, n_i being the number of sampling days which sample i contributed to each segment. The mean concentration $\overline{C}$ in each segment was estimated from

$$\overline{C} = \frac{\sum n_i C_i}{\sum n_i}.$$

In the same fashion, weighted standard deviations were calculated. The long sampling time inevitably smoothed the true variability of concentrations, but since one month was a typical sampling length the result is comparable to the monthly averages published for other tracers. On the average, each of the 25 segments was covered 6.8 times by the total data set.

Fig. 1 shows the mean annual concentration cycle for EC at Cape Grim. The variance of the data is indicated by a shaded band of plus/minus one weighted standard deviation about the weighted mean. The weighted standard deviation in each segment is quite high, on the average 51% of the mean. Near the main annual peak, standard deviations up to 80% occurred. Relative experimental errors are far smaller (about 20% of the mean). There is a large interannual variability, as one would expect from an incompletely mixed tracer. This of course, will reduce the value of a comparison with tracers not measured over the same sampling period. However, a simple statistical test reveals that the main features of the average seasonal variation of EC are significant. The difference between the four consecutive segments with highest concentrations and the four consecutive segments with lowest concentrations is highly significant at the 0.5% level. A second mode in the annual cycle of EC can be distinguished in the period April–May. Because of the fact that it is only supported by data of four years out of eight and because of the generally

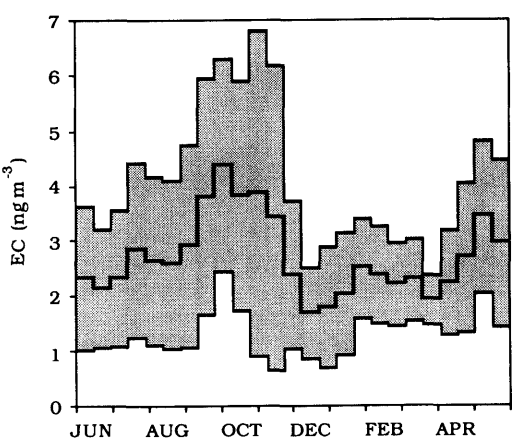

Fig. 1. Mean seasonal cycle in elemental carbon, (EC), at Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E). The shaded band about the mean indicates one standard deviation.

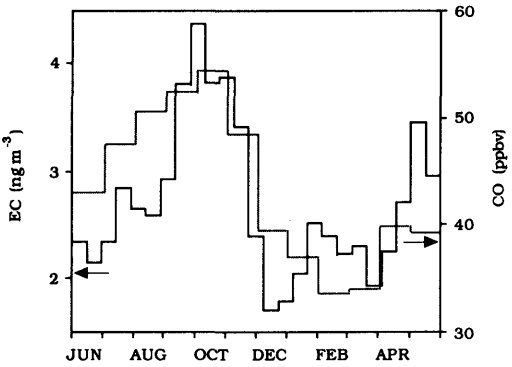

Fig. 2. Comparison of the seasonal cycle in EC and carbon monoxide, (CO), from Fraser et al. (1987) and Fraser et al. (1988) at Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E).

high variance we refrain from a detailed analysis of this secondary feature at this point.

In Fig. 2, published monthly averages (Fraser et al., 1987 and Fraser et al., 1988) for CO for 1982–1986 are compared with the EC curve. In spite of the above considerations, the shapes of the curves are surprisingly close. The scales have been adjusted to emphasize the similarities and to disguise the fact that the amplitude of the CO variation is about $\pm 30\%$ of the mean while the corresponding value for EC is $\pm 40\%$; (the difference would be reduced slightly if monthly averages had been used for EC). Both seasonal curves show a large October maximum with a sharp decrease to a broad summer minimum. The most important relative differences are lower EC values in winter (June, July, August) and lower CO values from February to May.

3. Possible causes of the seasonal variation

The close similarity in the main features of the seasonal cycles of both EC and CO suggests a common cause of the changes in both. The differing loss mechanisms—scavenging by precipitation for EC and reaction with OH for CO—appear to be of minor importance. Scavenging will be most effective in June, July and August at Cape Grim, where winter cloudiness and precipitation are a maximum. However, the baseline sampling conditions might have suppressed part of the seasonality in the scavenging affecting the EC-data, in particular in the vicinity of the site. EC values at this time are low relative to those of CO. The relatively low CO values from February to May might be due to enhanced OH concentrations, though there is no evidence available to support this.

Clearly, we have to seek the main causes of the seasonal cycles elsewhere. Some possible controls on concentration changes in these two substances are changes in:

(1) lower tropospheric transport in the southern hemisphere;
(2) mixing with the upper troposphere;
(3) interhemispheric transport;
(4) seasonal variation in source strength.

To examine the first three controls more in detail, we will look at the annual cycle of other tracers at Cape Grim and their implication for the seasonal changes in EC. An investigation of the fourth control will lead us to look at tracer measurements from other southern hemisphere sites. Thus, we shall dedicate several subsequent sections to its discussion.

3.1. Radon as a tracer of ground level continental air

Radon, ^{222}Rn, is one of the radioactive decay products of radium, is produced in appreciable quantities only over ice-free land, does not react with any other atmospheric constituent and is not subjected to atmospheric scavenging processes. With its half-life of only 3.8 days it is ideally suited to detecting changes in the transport of air from the nearest continents. Beyond Australia the next continent upwind is Africa, no part of which lies less than about 6° north of Cape Grim. In the area south of Cape Horn, Schwerdtfeger (1962) has shown the existence of "equinoctial storms" in terms of fall and spring maxima of the westerly wind component from the surface to the 500 hPa level. Therefore we shall look if this semi-annual variation in zonal transport is reflected in the radon concentrations at Cape Grim.

Monthly mean ^{222}Rn concentrations for the four years 1983–1986 have been derived from the list of monthly means under baseline conditions published by Whittlestone et al. (1988). These are compared in Fig. 3 with the EC variations of Fig. 2. The two seasonal cycles are out of phase, radon showing a minimum during the period of the

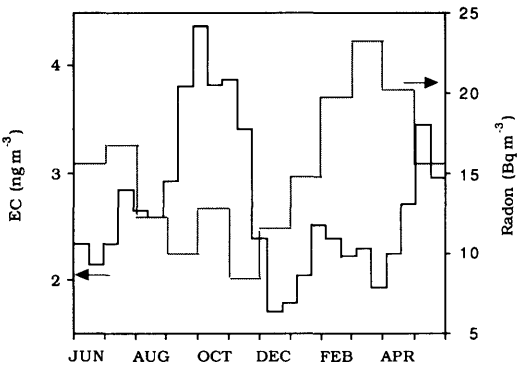

Fig. 3. Comparison of the seasonal cycle in EC and ^{222}Rn (from Whittlestone et al., 1988) at Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E).

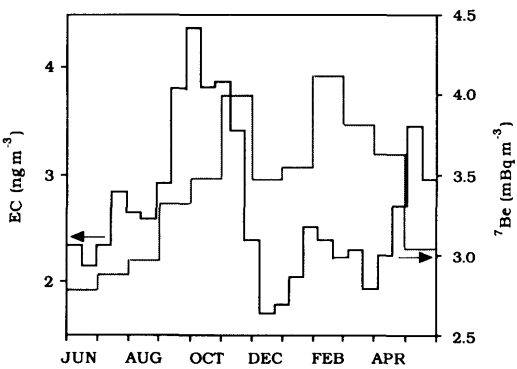

Fig. 4. Comparison of the seasonal cycle in ^{7}Be (from Prospero, 1987 and Larsen and Prospero, 1988) and EC at Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E).

spring maximum in EC and maximum in March when EC concentrations are low.

We can therefore rule out accelerated southward transport from continental source regions as a cause of the spring maximum in EC- and CO concentrations. However, radon concentrations show a clear maximum during the fall period of enhanced zonal transport in the southern hemisphere and remain high until the end of May. Thus aged continental emissions might contribute to the steep rise in EC concentrations in April and to the short-lived maximum in May.

3.2. Mixing with the upper troposphere

Prospero (1987) and Larsen and Prospero (1988) have reported monthly means of ^{7}Be at Cape Grim. This tracer is a radionuclide produced mainly in the upper troposphere by cosmic ray bombardment and having a half-life of 54.5 days. Its concentration is therefore a measure of mixing with the upper troposphere over a wide area centered on the position of the sampling site. Monthly mean concentrations of ^{7}Be are compared in Fig. 4 with EC concentrations. There is such a large difference that it must be concluded that downward mixing of EC transported at higher levels in the atmosphere does not provide an important contribution to the spring maximum.

3.3. Interhemispheric transport

Enting (1987) has shown that the seasonal cycle of CO_2 concentrations both at Cape Grim and Mauna Loa, Hawaii rule out appreciable interhemispheric transport. We investigated interhemispheric transport by looking at the seasonal variability at Cape Grim of any chlorofluorocarbon which is overwhelmingly produced in the northern hemisphere. For example, if the trend is extracted from the CCl_2F_2 measurements reported by Fraser and Derek (1987, 1988) and means taken for the years 1982–1986, the months April to August have means 0.5% higher than the other months (see Fig. 5). This period precedes the peak in tracers which we suspect to be combustion derived at Cape Grim by two months. If the CCl_2F_2 maximum is due to interhemispheric transport, northern hemispheric non-biomass combustion sources might contribute a minor fraction to the spring maximum in EC, CO and CH_4. We shall see below that northern hemispheric tropical fire emissions and methane emissions from wetlands peak during the period of CCl_2F_2 decrease towards the low spring and summer concentrations. This phase shift would explain why sources in the northern hemisphere are not influencing EC, CO and CH_4 significantly at Cape Grim.

Another tracer measured at Cape Grim is ^{210}Pb, the end product of radon decay. Although this isotope has the very long half-life of 22 years, its attachment to submicron particles makes it susceptible to loss by tropospheric scavenging processes. Even though only a limited amount of radon can diffuse into the upper troposphere or lower stratosphere, the greatly increased resi-

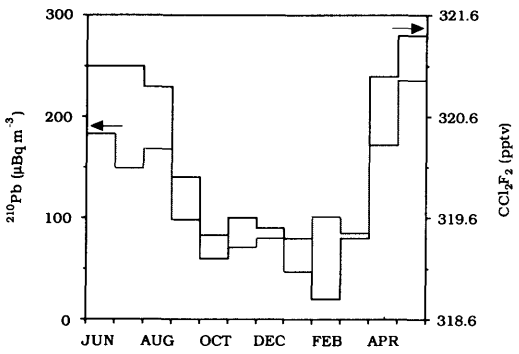

Fig. 5. Average seasonal cycle of CCl_2F_2 (from Fraser and Derek, 1987, 1988) and ^{210}Pb (from Prospero, 1987 and Larsen and Prospero, 1988) at Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E) for the years 1982–1986. The trend has been removed from the CCl_2F_2 data.

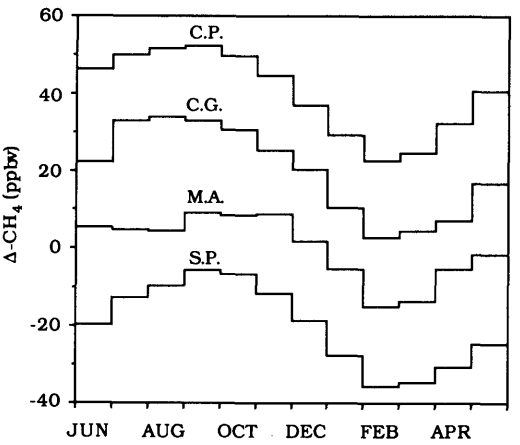

Fig. 6. Comparison of the annual cycles in methane at Cape Point, South Africa (lat. 34°S, long. 18°E) (from Brunke et al., 1990), Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E) (from Fraser et al., 1987, 1988), Mawson, Antarctica (lat. 68°S, long. 63°E), and South Pole (the latter two from Fraser et al., 1986). The data are given in deviations from the mean values. For clarity of presentation the cycles C.P., C.G., M.A., and S.P. have been offset by 40, 02, 0, and −20 ppbv respectively.

dence time of the resulting ^{210}Pb at these levels makes interpretation of concentration changes difficult. Like CCl_2F_2 its maximum values occur from April to August (see Fig. 5). This result might also be consistent with high altitude interhemispheric transport.

4. Seasonal variation in source strength

4.1. Methane

We begin our discussion of the seasonality in source strength by adding methane (CH_4) concentrations to our considerations. While most of the methane production is estimated to be of microbial origin, 3–10% are due to biomass burning (Hao et al., 1989). Corresponding figures for CO are 11–36% (Hao et al., 1989). Fraser et al. (1986) have found that seasonal cycles in CO and CH_4 are very similar. This can be seen at several stations from Cape Point, South Africa to South Pole by comparing Figs. 6 and 7.

The main difference is the lower amplitude of the CH_4 cycle, which of course would be expected from its much longer residence time of about ten years. There is also some smoothing of the asymmetry of the CO curve. The great similarity implies that there is no marked seasonality in non-biomass burning sources of methane in the southern hemisphere. Emissions

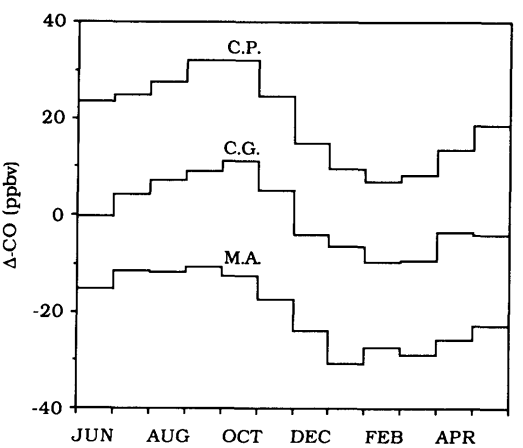

Fig. 7. Comparison of the annual cycle in CO at Cape Point (lat. 34°S, long. 18°E), Cape Grim, Tasmania (lat. 40.7°S, long. 144.7°E) and Mawson, Antarctica (lat. 68°S, long. 63°E). The data are given in deviations from the mean values. For clarity of presentation the cycles C.P., C.G., and S.P. have been offset by 20, 0, and −20 ppbv, respectively.

from natural fresh water wetlands and rice paddies are two of the major sources of methane. Aselmann and Crutzen (1989) have shown that (a) the major regions for these sources are in the northern hemisphere and (b) that both sources exhibit their (annual) minimum emissions in the southern hemisphere during the months July–October.

Hao et al. (1989) have made estimates of trace gas emissions from tropical fires based on the FAO survey for the period 1975–1980. They used monthly rainfall statistics to estimate the seasonal variation of emissions. They assumed a symmetrical distribution with the weights 1:2:2:2:1 of biomass burning during the last five months of the dry season. Their results in 5° × 5° cells have been used to calculate an approximate seasonal variation in emissions from biomass burning in the tropical regions of the northern and southern hemispheres. In Fig. 8 this variation is plotted for CO_2 emissions. According to Crutzen et al. (1985), CO and CH_4 emissions can be calculated from the CO_2 emissions by applying the volumetric emission ratios 0.12–0.15 and 0.008–0.016 respectively.

Estimated maximum emissions in the southern hemisphere precede maximum concentrations in EC, CO and CH_4 at Cape Grim by two months. While northern hemispheric emissions are on the same order of magnitude as in the southern hemisphere their peak in January–April is not reflected in the trace gas data at Cape Grim.

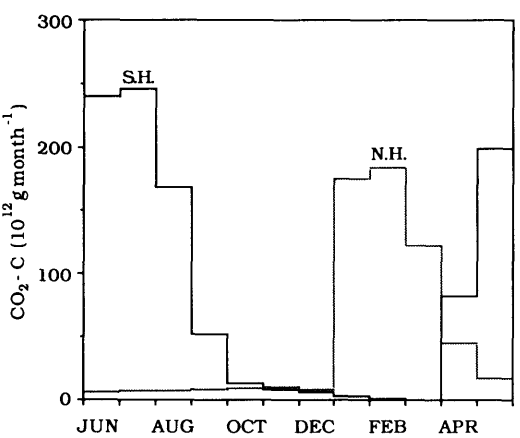

Fig. 8. Seasonal variation in CO_2 emissions from biomass burning in the tropical regions of the southern (S.H.) and northern hemisphere (N.H.), after Hao et al. (1989).

4.2. Carbon monoxide

In order to shed light on the apparent discrepancy between the seasonal cycle of emissions from biomass burning and that of combustion tracers at Cape Grim we will look at results from other southern hemispheric monitoring sites. At Cape Point, South Africa (lat. 34°S, long. 18°E), CO has been measured since 1978. The most recent average seasonal cycle was reported in Brunke et al. (1990). At Mawson, Antarctica (lat. 68°S, long. 63°E), CO has been measured since 1979 and an average annual cycle has been published in Fraser et al. (1986). There is a good agreement in seasonal variation with the corresponding data from Cape Grim (see Fig. 7). Seiler et al. (1984) explained the annual CO cycle at Cape Point by a seasonal variation of OH concentrations and by the seasonal variation of the continental source area south of the ITCZ exposed to rapid intrahemispheric mixing. As with EC the concentration build up in CO towards the September/October peak is more gradual than the decrease towards summer.

A recent investigation of biomass burning also indicates strong emissions at the end of the dry season. According to Andreae (private communication) high levels of CO, O_3, particle number and clearly visible haze layers in September 1989 in Natal, Brazil could be traced back to the African continent.

4.3. Particulate tracers at other southern hemispheric sites

Seasonal variation of particulate tracers at southern hemispheric sites are very scarce. Some evidence for maximum combustion emissions towards the end of the dry season can be found in the data on aerosol optical depth measured at Keetmanshoop, South Africa (26.5°S, 18°E) which are reported in WMO (1974–1977). During 1972–1975, sun photometer measurements were made intermittently at that site. The average annual cycle of aerosol turbidity at 0.5 μm wavelength has its peak in September. However, the decline to the summer months is more gradual than that of the tracers at Cape Point and Cape Grim. At the same time, we have to allow

for an alternative explanation of the aerosol data due to the seasonal variation of soil dust production.

Aerosol measurements have been performed at several coastal sites of Antarctica and at the South Pole. In most cases the total number of particles has been measured which is dominated by regional production sources (Shaw, 1988). Information on particles in the size range which is most important in long range transport of air pollutants, (0.1–several μm), has been derived mostly from measurements with integrating nephelometers (Bodhaine and Bortniak, 1981), and in a few cases from particle counters (Ito, 1989). In the size range >0.15 μm radius Ono et al. (1981) found a concentration maximum in September 1978 at the Station Syowa (69°S, 39.6°E) which they could not explain at that time. Later Ito (1985) explained this maximum by a maximum influx of marine air which brings high concentrations of "maritime background aerosols" in this month.

At the South Pole, the average annual variation of nephelometer results shows a minimum in May and a double maximum in the period August–October. At the time of the annual maximum the spectral distribution of the nephelometer data indicates the presence of larger particles than during the Austral summer. From this fact together with the results of chemical analyses on concurrent filter samples it has been concluded that the intrusion of warm sea salt laden air masses is the cause of the seasonal variation of particles larger than ca. 0.1 μm over Antarctica (Bodhaine et al., 1987). However, no direct relationship between particulate mass, nephelometer signals and sodium concentrations (for sea salt) over Antarctica has been established yet.

To date, the only direct information on particulate combustion products over Antarctica is a one-year time series of EC concentrations taken at the South Pole (Hansen et al., 1988). The monthly averages at the South Pole and at Cape Grim have quite similar seasonal variations. At the South Pole the large peak followed by a steep decline occurred two months later than at Cape Grim. If this similarity is not fortuitous, it would suggest that the cause of the maximum in both curves is simply the southward progression of air masses polluted by biomass burning at lower latitudes. A direct correlation of EC results and nephelometer data and sodium concentrations ought to reveal how much of the spring peak in particulate light scattering is due to sea salt and how much due to the transport of biomass burning products in the same air masses coming from lower latitudes.

5. Conclusions

The tentative interpretation of this array of seasonal cycles of different tracers at southern hemispheric sites between 26°S and 90°S is that EC, CO and CH_4 owe most of their seasonal variability to a peak in production in areas with biomass burning in the southern hemisphere. Combustion products from these regions are mixed rapidly throughout the southern hemisphere. If so, it is perhaps fortunate for the cleanliness of the Antarctic that the production does not occur at the time of maximum poleward transport but instead at its minimum. None of the other possible processes controlling seasonality seems capable of explaining more than minor features of the seasonal cycle.

Perhaps the most important conclusion is that it seems unlikely that there is a substantial seasonal variability in OH which has previously been thought to have a major influence on the annual cycles of CO and CH_4 in the Antarctic.

However, some doubts remain on the validity of the simple concept of southward-moving pollutants from biomass burning. These are mainly due to the two-month discrepancy between the traditional view of the biomass burning season and the peak of the concentrations of combustion products in the southern hemisphere. To remove these doubts, simultaneous measurements of EC with high time resolution at several sites—for example Cape Point, Cape Grim, Macquarie Island (54.5°S), a coastal Antarctic site (about 67°S) and the South Pole would eventually show the reality of our hypothesis. At the same time, such measurements in conjunction with those of the other substances discussed in this paper would provide valuable insights into atmospheric transport. Such information seems essential to a full understanding of changing concentrations of many trace substances and their environmental effects.

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