Tellus (2001), 53B, 10–21
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TELLUS
ISSN 0280–6509

Anthropogenic and biogenic winter sources of Arctic CO_2 — a model study

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(Manuscript received 18 January 1999; in final form 30 May 2000)

ABSTRACT

Long-range transport of anthropogenic and biogenic CO_2 to a remote site in the Arctic is studied. A limited area, off-line, Eulerian atmospheric transport model is used, and the results are compared to the observed CO_2 concentration at the "Ny-Ålesund International Arctic Research and Monitoring Facility". Inventories of anthropogenic CO_2 emissions and estimates of biogenic CO_2 emissions are used to investigate the respective impact of these emissions on Arctic CO_2 variations during 4 winter months. A direct comparison of the modelled and observed concentrations reveals remarkably good timing in the modelled variations as compared to the observed variations for most of the time. The correlation of observed versus modelled CO_2 concentration is significant at the 95% confidence level. The biogenic and the anthropogenic CO_2 emissions are shown to have approximately equal influence on Arctic CO_2 variations during winter. Europe is found to be the dominant source of anthropogenic CO_2 at the monitoring station, while Siberia and Northern America have little influence on Arctic CO_2, during the months studied. These results contradict Engardt and Holmén whose results indicate that the lower-Ob region in western Siberia has a large impact on Arctic CO_2.

1. Introduction

To increase the understanding of long-range transport of polluted air to the Arctic, a number of monitoring stations have been set up. Measurements of atmospheric CO_2 in the Arctic are performed at Alert, Canada (Wong et al., 1984; Higuchi and Daggupaty, 1985), Barrow, Alaska (Peterson et al., 1982) and at Ny-Ålesund, Spitsbergen (Holmén et al., 1995). The variations of atmospheric CO_2 on the synoptic time scale are determined by local/regional sources, but also by long-range transport from distant sources. We present a comparison between modelled and observed variations of CO_2 concentrations on the synoptic timescale for the monitoring station at Zeppelinfjellet, Ny-Ålesund, Spitsbergen. This enables a study of the spatial distribution and absolute magnitude of the anthropogenic and biogenic emissions of CO_2 to the atmosphere.

This study is a continuation of the work performed by Engardt and Holmén (1999, henceforth EH99). The model used is an Eulerian 3-dimensional atmospheric transport model, which utilizes analysed meteorological data from the European Centre of Medium-Range Forecasts (ECMWF). The model is described in EH99 and Robertson et al. (1999). In EH99 this model is used to determine which anthropogenic sources are important for the observed CO_2 at Zeppelinfjellet. They find that the industrial centres and surrounding gas-fields in the lower-Ob region (60°–72°N, 65°–80°E) in western Siberia often have a large impact on the modelled CO_2. A high correlation between the aerosol particle scattering coefficient measured at Zeppelinfjellet

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and the modelled CO_2 is also found. Engardt and Holmén do therefore not consider it likely that biogenic CO_2 emissions are a significant source for the observed CO_2 at Zeppelinfjellet during the two winter months they study. To complement the two months studied in EH99 and to increase the credibility of the results, four different winter months are chosen here to represent different origins of the air reaching the Zeppelinfjellet monitoring station.

The amplitude of the modelled CO_2 variations is consistently too low in EH99, as compared to the observations, and two alternative explanations for this discrepancy are explored. One possible explanation is weaknesses in the anthropogenic emission inventory, such as the omission of (possibly) large emissions in the lower-Ob region. Since Engardt and Holmén performed their study, a new inventory of anthropogenic CO_2 emissions (Olivier et al., 1996) has been compiled. This new, more detailed, inventory includes emissions from the gas-fields in the lower-Ob region and is used in the present study.

Another possible explanation for the low amplitude variations is the omission of biogenic CO_2 emissions. Zondervan and Meijer (1996) analyse a positive anomaly in the wintertime CO_2 record at Kollumerwaard, the Netherlands. Based on simultaneous ^{14}C measurements to determine the biogenic influence on the observed variations in CO_2, they find that the anthropogenic and biogenic influences are *correlated*. Further, Yuen et al. (1996) find that biogenic emissions of CO_2 have a larger impact than anthropogenic emissions on an anomaly in the record of atmospheric CO_2 in December at Alert, Canada. Yuen et al. use an atmospheric transport model similar to the model used in the present study. Following on both these results we investigate the effects of biogenic CO_2 emissions on the variations in atmospheric CO_2 at Zeppelinfjellet during winter.

The horizontal and vertical resolution may also cause some disagreement between the model results and the CO_2 data, but this is not further explored in this study. The sources of CO_2, both anthropogenic and biogenic, vary on a synoptic and diurnal time scale, but the flux data used in this study are only available as monthly means. A discussion of the influence of these variations on the variations of CO_2 at Zeppelinfjellet can therefore not be pursued.

In Section 2 we describe the differences in the model setup as compared to the EH99 study. The four winter months we study are described in Section 3 in terms of the CO_2 record at Zeppelinfjellet and five-day back-trajectories. We present the results of the study in Section 4 and the conclusions in Section 5.

2. Current set-up

In EH99, it is assumed that the influence of the atmosphere-biosphere exchange on wintertime Arctic CO_2 is negligible. However, both modeling results (Yuen et al., 1996; Bonan, 1996) and measurements (see Subsection 2.2), indicate that these fluxes can be of importance also during winter months. We therefore investigate the influence of biogenic CO_2 emissions on Arctic CO_2 concentrations. We continue to assume that the net flux of CO_2 between the northern North Atlantic and the atmosphere is negligible.

In EH99, a series of tests are performed where the model is initialised with different initial and boundary conditions for January and February 1993. They conclude that the background mixing ratio is determined by the amount of CO_2 at the model boundaries, while the synoptic variability is generated by sources within the model domain. Following on their results we start the integrations with zero CO_2 in the model domain and zero influx through the boundaries.

2.1. *Anthropogenic source functions*

We compare the synoptic variations in CO_2 at Zeppelinfjellet modelled using three different anthropogenic source functions. Some characteristics of the source functions are listed in Table 1.

In EH99, an inventory of anthropogenic CO_2 emissions compiled by Andres et al. (1996) (henceforth A96) is used. Since Engardt and Holmén performed their study a new database for anthropogenic CO_2 emissions has been compiled, the EDGAR V2.0 database (Olivier et al., 1996, henceforth EDGAR). Both A96 and EDGAR describe a global 1° latitude $\times 1^\circ$ longitude map of anthropogenic CO_2 emissions valid for 1990. However, EDGAR has the emissions distributed by the actual locations of the sources, while A96 distribute the emissions based on population density.

Table 1. *A short description of the different anthropogenic and biogenic source functions used in the study; the total CO_2 emissions presented for the biogenic source functions are valid for the month of December*

Source function	Description	Total CO_2 emissions in the model domain (Pg C yr^{-1})
Anthropogenic		
Andres et al. (1996)	National fossil fuel CO_2 emissions spread within each country based on population density	1.94
EDGAR database	CO_2 emissions distributed based on knowledge of the actual source locations. Also includes, e.g. biomass burning, oil production, transmission and handling	2.10
Benkovitz et al. (1996)	Global distribution of anthropogenic sulphur (SO_2 and SO; SO_x) emissions. Used in EH99, scaled linearly such that the total CO_2 emissions in the model domain are the same as in A96	1.94
Biogenic		
Fung et al. (1987)	Seasonal exchange of CO_2 between the atmosphere and the terrestrial biosphere derived using satellite measurements of radiance and field data of soil respiration. Used by Yuen et al. (1996)	1.96
CCM3	Surface biospheric fluxes calculated in the NCAR CCM3, a full GCM coupled to an advanced land surface parameterisation	2.38

Further, EDGAR includes emissions from oil production, transmission and handling, which are not included in A96. This allows a test of the hypothesis in EH99 of the gas fields in the lower Ob-region as a source of elevated Arctic CO_2 concentrations.

In order to investigate the influence of the geographical distribution of the emissions in the A96 inventory on the variations in CO_2 at Zeppelinfjellet, an alternative anthropogenic source function is used in EH99. It is obtained by a linear scaling of a global distribution of anthropogenic sulphur (sulphur dioxide and sulphate; SO_x) emissions compiled by Benkovitz et al. (1996, henceforth B96). The scaling is such that the total CO_2 emissions within the model domain are the same as in the A96 inventory. Hence,

$$\mathrm{EMIS}_{\mathrm{CO_2}} = \mathrm{EMIS}_{\mathrm{SO}_x} \cdot \frac{\mathrm{TOT}_{\mathrm{CO_2}}}{\mathrm{TOT}_{\mathrm{SO}_x}}, \quad (1)$$

where $\mathrm{EMIS}_{\mathrm{CO_2}}$ and $\mathrm{EMIS}_{\mathrm{SO}_x}$ are the emissions at a grid point and $\mathrm{TOT}_{\mathrm{CO_2}}$ and $\mathrm{TOT}_{\mathrm{SO}_x}$ are the total emissions in the model domain for CO_2 and SO_x respectively. The CO_2 emissions in the Ob-Norilsk region given by this scaling are almost ten times larger than the emissions in this region in EDGAR.

2.2. *Biogenic source functions*

We also compare the synoptic variations in CO_2 at Zeppelinfjellet modelled using two different biogenic source functions. Some characteristics of the source functions and the total emissions in the model domain for the month of December are listed in Table 1. Note that the total biogenic emissions of CO_2 are of the same order as the anthropogenic emissions in the model domain in winter.

Measurements of the rates of photosynthesis, respiration and decomposition made throughout the year and over large areas are sparse. However, Fung et al. (1987) (henceforth F87) derived the seasonal exchange of CO_2 between the atmosphere and the terrestrial biosphere using satellite measurements of radiance and field data of soil respiration. This exchange distribution was used by Yuen et al. (1996) to simulate the large positive anomaly of atmospheric CO_2 over the Canadian Arctic mentioned in Section 1.

Also used in the present study are the monthly mean surface biospheric fluxes (SBF) calculated in CCM3, the National Center for Atmospheric Research Community Climate Model. CCM3 is a full atmospheric GCM coupled to an advanced land surface parameterisation (Bonan, 1996). The

horizontal distribution of the net flux from the terrestrial biosphere in December from the CCM3 is shown in Fig. 1. The horizontal distribution of the net flux in F87 differs from that of the CCM3 field by showing larger amplitude maxima in Europe and a zero efflux in Siberia for December.

For the present study, calculated fluxes from the CCM3 are only available for the months October through January. To study the month of February 1993 we have therefore used SBF from the CCM3 for *January* instead. We have compared two runs for February 1993 with December and January SBF respectively. Based on these runs we estimate the maximum overestimation of the amplitude of the modelled variations in CO_2 at Zeppelinfjellet, introduced when inadequate fluxes are used for this month, to $\approx 15\%$.

The SBF in CCM3 and F87 have been compared to measurements of the winter CO_2 flux from the surface. Zimov et al. (1993; 1996) measure the wintertime CO_2 emissions from soils in north-eastern Siberia and estimate the average CO_2 flux from soils to be 0.26 g C m^{-2} day^{-1} for December. This value is more than ten times larger than the flux given by the CCM3 field for this area. Dörr and Münnich (1987) measure the CO_2 production

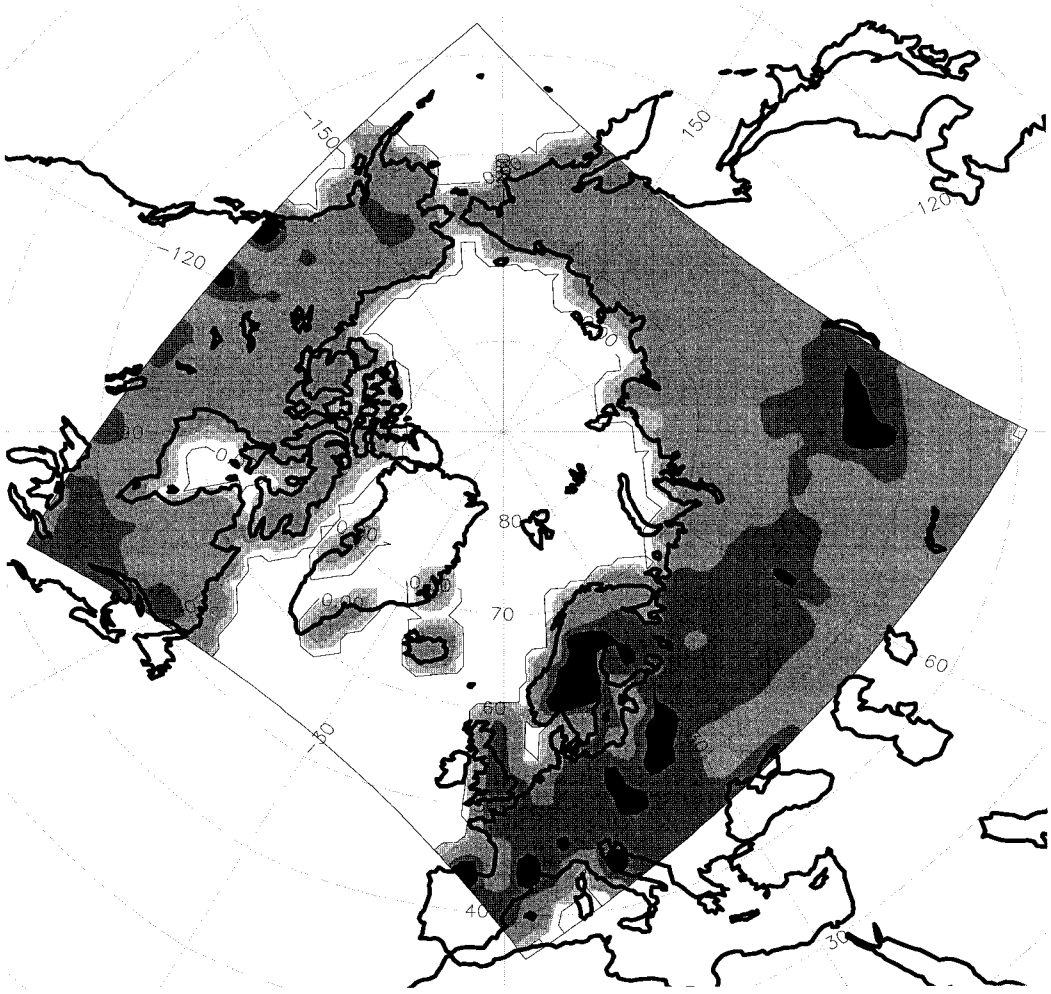

Fig. 1. Biogenic CO_2 emissions from the CCM3 for the month of December (Bonan, 1996). Units are g C m^{-2} day^{-1} and light grey is 0.001–0.25, darker grey is 0.25–0.50 and black is 0.50–1.00.

in soils at 35 locations with different soils and vegetation in an area of about 50 kilometers radius around Heidelberg in the Rhine valley. They find an average CO_2 production for November through February of 3 g C m^{-2} day^{-1}. The same winter flux is found by Schmidt et al. (1996), who use measurements of ^{222}Rn and CO_2 to derive an average winter biogenic flux of CO_2 for Germany. This flux is three times larger than the flux in this area given by CCM3 and F87.

Surface fluxes can be influenced by synoptic scale weather variations but such effects are not believed to influence the present results.

2.3. *Geographical source regions*

The model is run with the anthropogenic and biogenic source functions described above. The EDGAR source function is also divided into geographical source regions, to determine the origin of the modelled anthropogenic CO_2. These regions are Europe (30°W–60°E), western Europe (30°W–25°E), Siberia (60°–180°E) and the Ob-Norilsk region (60°–72°N, 65°–90°E).

3. Four winter months

3.1. *CO_2data*

The measurements of atmospheric CO_2 used here were performed at Zeppelinfjellet, a mountain ridge a few km south of Ny-Ålesund, Svalbard. The station is situated approximately 1000 kilometers from the nearest human activities of size, the only local sources being 3–4 Norwegian and Russian settlements about 100 kilometers south of the station. As most of Spitsbergen is covered with glaciers and the vegetation in the fjord valleys is sparse, all hours and wind-directions are considered representative of background conditions.

The concentration (mixing ratio) of CO_2 has been measured at Zeppelinfjellet since 1990, using an UNOR 4N instrument utilizing non-dispersive infra-red technique (Holmén et al., 1995; Engardt et al., 1996). Since February 1994 weekly samples are also collected at Zeppelinfjellet and analysed at the NOAA/CMDL in Boulder, Colorado. Comparisons between the continuous data from the Zeppelinfjellet station and the NOAA/CMDL data show correspondence within ± 0.3 ppmv (Holmén et al., 1995).

As noted in EH99, the background CO_2 concentration at the monitoring station is determined by the amount of CO_2 at the model domain boundaries. We perform our calculations with zero influx through the boundaries in order to study the synoptic variations generated by sources within the model domain. Hence, in order to compare the modelled and the observed concentrations of CO_2, we perform a least-square fitting of the consecutive 5-day mean observed CO_2 concentration to a linearly increasing harmonic function,

$$q(t) = q_0 + q_1 t + \sum (a_k \sin 2\pi kt + b_k \cos 2\pi kt), \quad (2)$$

where $k = 1$ to 4, t is time and q is mixing ratio of CO_2. q_0, q_1, a_k and b_k are the coefficients to be fitted. We perform this fitting of the observed CO_2 mixing ratio from 1989 to 1998 and subtract this background variation from the observed CO_2 at Zeppelinfjellet. We also add a constant value to the timeseries of observed CO_2 for each month in order to obtain non-negative values of the mixing ratio. The concentration of CO_2 thus obtained (henceforth C_o) is used to evaluate the model results.

3.2. *Trajectories*

In this study, use is made of 3-dimensional trajectories calculated at the ECMWF. They have been calculated using the R. McGrath (McGrath, 1989) model which is based on the ECMWF analysed wind and pressure fields and a calculated vertical wind. Five-day trajectories arriving at the location of Zeppelinfjellet (79.9°N, 11.9°E) at 950 hPa are used to provide an indication of the source of the air arriving at the given location.

3.3. *Case descriptions*

Four months (February, November and December 1993 and January 1995) are chosen so that they represent different origins of the air reaching the Zeppelinfjellet monitoring station. The choice is based on trajectories and C_o at Zeppelinfjellet for the three winters (November through February) 1992–93, 1993–94 and 1994–95. A plot of 5-day back-trajectories arriving to Zeppelinfjellet at 12 UTC is shown in Fig. 2. These trajectories give an overall picture of the origin of the air arriving at Zeppelinfjellet during the

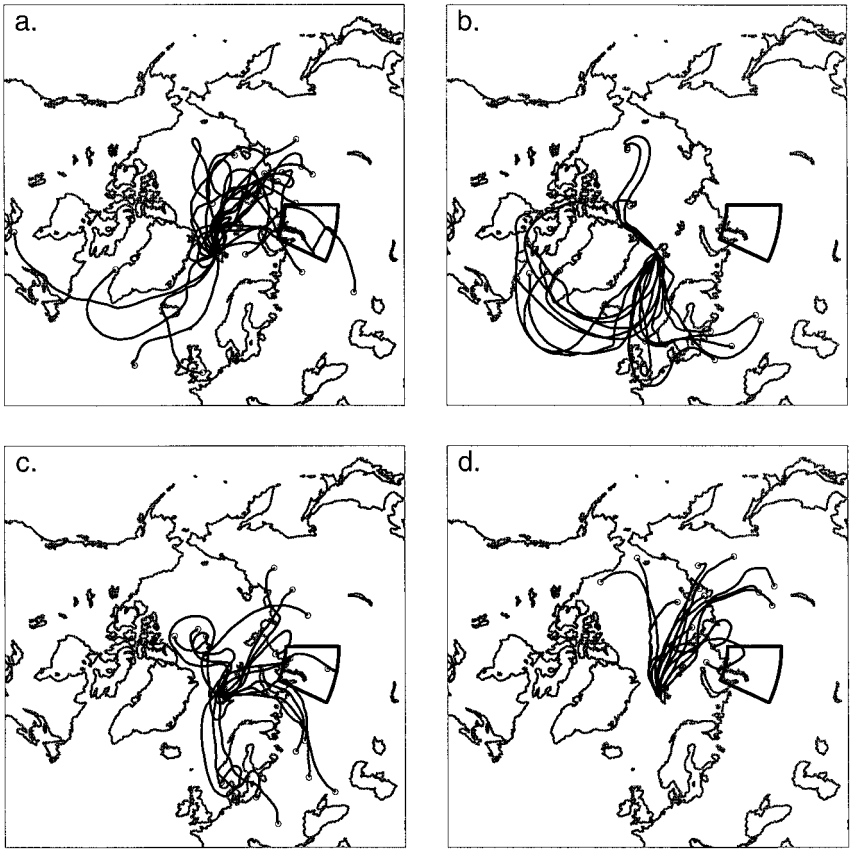

Fig. 2. One trajectory for each day of the month arriving at 12.00 a.m. UTC at Zeppelinfjellet for (a) February 1993, (b) November 1993, (c) December 1993 and (d) January 1995. Trajectories which evidently have entered a cyclone or a frontal zone (i.e., exhibit 180° turns or small-radii circling motions) during the 5 days are omitted. The lower-Ob region is indicated by a box in each figure.

months in question. The lower-Ob region is also indicated in the figure. A short motivation of the choice of each of the four months is given in the following.

February 1993 is discussed in EH99 and a further investigation of this month is performed here. The trajectories for this month indicate a preference for transport from Siberia, with some transport over the North Atlantic Sea and also passing the lower-Ob region.

November 1993 differs from the other four months compared, both in C_o and in the origin of the air reaching Zeppelinfjellet. C_o shows narrow peaks with events lasting from hours to a day or two. This is in contrast to C_o for all other winter months studied, which show broader peaks with pollution events lasting for several days. The collection of trajectories for November 1993 indicate transport directly from northern Europe, via the North Sea to Zeppelinfjellet or, transport from northern North America over the northern Atlantic to Zeppelinfjellet.

December 1993 is chosen in order to test the hypothesis of EH99 that the industrial centres and the surrounding gas-fields in the lower Ob-region have a large impact on the observed CO_2 at Zeppelinfjellet. The trajectories indicate some transport of air from the eastern parts of Europe passing this region.

January 1995 has smaller amplitude variations in C_o than during the three other months studied. The trajectories indicate slow transport from east-

ern Siberia and the Far East to Zeppelinfjellet during this month. This month is of interest since the air has not passed Europe immediately prior to traversing Siberia.

4. Results

The total modelled concentration of CO_2 at Zeppelinfjellet (C_m), produced by both anthropogenic and biogenic emissions, is shown in Fig. 3. The results presented here are obtained with anthropogenic emissions from EDGAR and biogenic emissions from CCM3. In Subsection 4.2 we discuss the results obtained with anthropogenic emissions from A96 and B96 and with biogenic emissions from F87.

Fig. 3 also shows C_o at Zeppelinfjellet. Before discussing the implications of the model results for the sources of the CO_2 measured at Zeppelinfjellet, we present a comparison of the model results with the observations.

4.1. Comparison of model results and observations

To assess the performance of the model we need to include both a scientific and a statistical component in the model evaluation study (Hanna, 1994). The scientific component includes evaluation of the model components and of the performance in general, which is performed by Robertson et al. (1999). It also includes a direct comparison of C_o with C_m. For the most part, the amplitude and timing of the variations in C_m is similar to the observed variations (Fig. 3). The modelled timing is at many instances remarkably good, while discrepancies in the amplitude are more frequent. Days when the difference between the modelled and the observed amplitude is very large are discussed in Subsection 4.3.

The hypothesis to be tested by the statistical evaluation is that there is no significant difference (at the 95% confidence level) between C_o and C_m for the four months we study. There is no universal measure that can be calculated to assess model performance. Hanna (1994) suggest a number of statistical parameters for the evaluation. Most of these parameters measure the difference between C_o and C_m. Due to the somewhat arbitrary way in which we set the zero for C_o (see Subsection 3.1), these parameters can not be used here. The following parameters are calculated: the correlation coefficient, R, and the standard deviations of C_o and C_m, σ_o and σ_m.

The correlation analysis is based on the assumption that the two datasets, C_o and C_m, contain independent data points. This is not true in our case, the concentration at one measurement time is highly correlated to the concentration at the next. Therefore, the degrees of freedom associated with the data set must be adjusted downward to reflect the actual number of independent points in the dataset. We have chosen to determine the number of independent points in the timeseries of C_o as the number of pollution events with an amplitude that exceeds 2 ppmv at some instant. This, somewhat arbitrary, choice can be regarded as an underestimation of the number of degrees of freedom. It yields 5 degrees of freedom for February 1993, 13 for November 1993, 4 for December 1993 and 4 for January 1995. We test our hypothesis on the four months together for which we have 24 degrees of freedom. The correlation coefficient is $R = 0.68$, which is significant on the 95% confidence level. The statistic R^2, which is an estimate of the fraction of the variance in C_o explained by the correlation, is $R^2 = 0.46$. This rather low value reflects 1. the periods of large errors in the modelled amplitude and 2. the difficulty of the model to reproduce the right timing in the variations on short timescales (on the order of a day). We also find that $\sigma_m \simeq \sigma_o$ with $\sigma_o = 1.82$ ppmv and $\sigma_m = 2.30$ ppmv, which demonstrates the model skill.

4.2. Comparison of different source functions

Using A96 for the anthropogenic emissions does not alter the timing of the variations, as compared to using EDGAR, but it produces somewhat lower amplitudes ($\leqslant 10\%$). When the anthropogenic CO_2 emissions spatially scaled to the SO_x inventory (B96) are used, the timing and amplitude of the variations at Zeppelinfjellet is similar to the timing and amplitude obtained using EDGAR for most times. However, at certain times the amplitude of the variations is up to 4 times larger using the B96 emissions than using EDGAR. These large amplitude variations are produced by the large emissions in the lower-Ob region.

When the biogenic emissions are taken from F87 the timing of the variations in CO_2 at

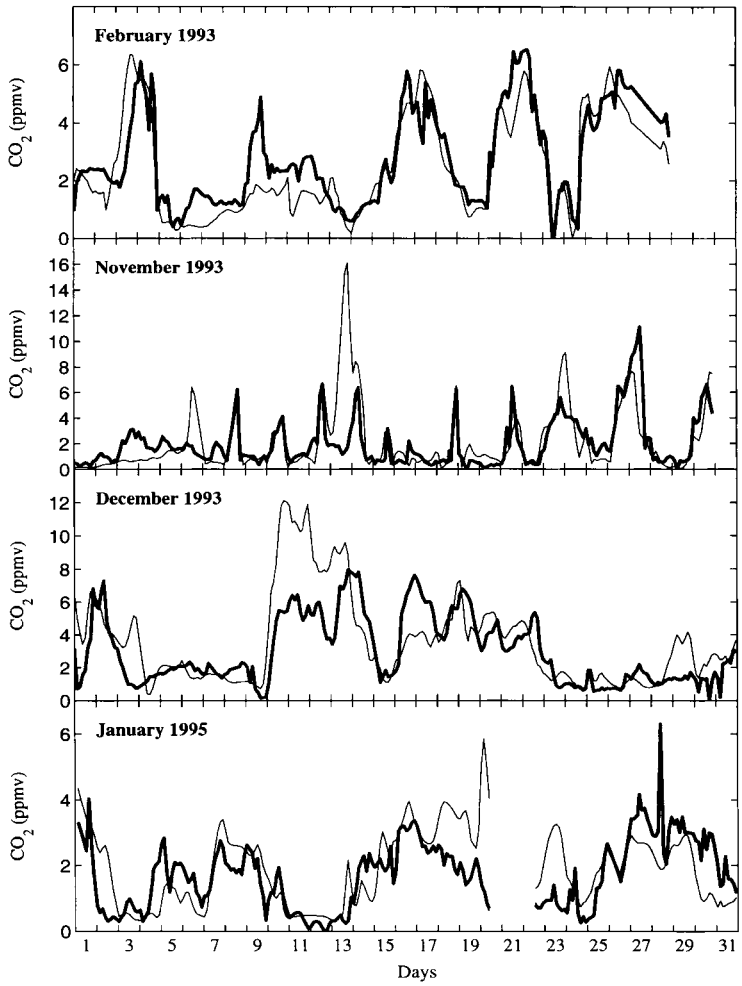

Fig. 3. The CO_2 concentration at Zeppelinfjellet during February, November and December 1993 and January 1995. The thick line shows the observed concentration and the thin line shows the total modelled concentration using both anthropogenic and biogenic emissions. The background variation of the observed concentration has been subtracted from the observed concentration. The curves of observed concentration are parallel shifted to obtain non-negative values of the concentration anomaly.

Zeppelinfjellet is less similar to the observed variations than when CCM3 biogenic emissions are used. The amplitude of the variations in CO_2 at Zeppelinfjellet when F87 is used is often twice that obtained with biogenic emissions from CCM3. The sum of the anthropogenic and biogenic emissions using biogenic emissions from F87 thus produces much larger amplitude variations than the observed variations. Hence, the results presented below utilize the CCM3 field.

4.3. *Sources of measured CO_2*

In order to determine the importance of different sources and geographical source regions for the variations in C_m at Zeppelinfjellet, we define the total amount of CO_2 reaching Zeppelinfjellet from both anthropogenic and biogenic emissions as

$$A_{\text{tot}} = \int (C_{\text{anthro}} + C_{\text{bio}})\, \mathrm{d}t, \tag{3}$$

Table 2. *The relative amount of CO_2 reaching Zeppelinfjellet from anthropogenic and biogenic sources in the model runs*

Source function	Region 1993	Feb. 1993	Nov. 1993	Dec. 1993	Jan. 1995
Biogenic (CCM3)	model domain	61%	42%	60%	60%
Anthropogenic (EDGAR)	model domain	39%	58%	40%	40%
	Europe	31%	56%	36%	34%
	Western Europe	14%	43%	18%	20%
	Siberia	7.7%	2.0%	4.8%	5.7%
	Ob-Norilsk	1.1%	<1%	<1%	<1%
Anthropogenic CO_2 scaled by SO_2 (B96)	Ob-Norilsk	11%	<1%	7.6%	11%

We define the total amount of CO_2 from both anthropogenic and biogenic sources as $A_{tot} = \int (C_{anthro} + C_{bio})\, dt$, where the integration is over one month and C_{anthro} and C_{bio} are the modelled concentrations at Zeppelinfjellet due to anthropogenic and biogenic emissions respectively. The relative amount of CO_2 from a particular source is now defined by $A_{rel} = \int C\, dt / A_{tot}$, where the integration again is taken over the complete month and C is the modelled concentration due to that particular source. Note that the relative amounts given for February 1993 are approximate due to the use of biospheric emissions valid for January.

where the integration is over the complete month and C_{anthro} and C_{bio} are the modelled concentrations at Zeppelinfjellet due to anthropogenic and biogenic emissions, respectively. The relative amount of CO_2 reaching Zeppelinfjellet from a particular source during 1 month is now defined as

$$A_{rel} = \frac{\int C\, dt}{A_{tot}}, \tag{4}$$

where the integration is, again, taken over the complete month and C is the modelled concentration of CO_2 at Zeppelinfjellet due to that particular source. The relative amounts of CO_2 of anthropogenic and biogenic origin are listed in Table 2 together with the relative amounts for the geographical source regions for anthropogenic emissions defined earlier. For comparison, the relative amount of CO_2 originating from the Ob-Norilsk region in B96, is also listed.

4.3.1. February 1993. Europe is the dominant anthropogenic source of the variations in C_m during February 1993 and the biogenic sources account for 61% of the amplitude of the variations in C_m.* As noted by EH99, the Ob-Norilsk region plays an important role for the variations in anthropogenic C_m during this month when the B96 emissions are used. However, using EDGAR for the anthropogenic CO_2 emissions, this region accounts for only $\approx 1\%$ of the total anthropogenic C_m. The correlation between the total C_m and the measured "aerosol concentration" during this month is discussed in Subsection 4.3.5.

4.3.2. November 1993. Europe, and in particular western Europe, is the major anthropogenic source of C_m during November 1993. The biogenic sources account for 42% of the amplitude of the variations in C_m. The modelled amplitude on the 6th and on the 13th of November is too large regardless of the source function used and the explanation for this discrepancy is likely to be found in the meteorological data and the model treatment of these.

4.3.3. December 1993. Europe is the major anthropogenic source during December 1993, with approximately equal parts from western and eastern Europe. The biogenic sources account for 60% of the amplitude of C_m. The amplitude of the variations in C_m between the 10 and 13 December 1993 is too large as compared to the observed amplitude, while the amplitude is too low between the 16 and 17 December. Part of the observed large amplitude during these days is reproduced when the B96 emissions are used. However, these

* Note that the value is too large ($\leqslant 15\%$) due to the use of January biospheric fluxes.

anthropogenic emissions also produce other large amplitude events during December 1993 with no correspondence in the observations. Hence, CO_2 emissions with a horizontal distribution coincident with the SO_x emissions are not supported by our model. We test if the emissions in EDGAR are too low in this region by increasing the emissions in EDGAR for the Ob-Norilsk region (to twenty times it's original value). This gives a better correspondence between the modelled and the observed amplitude during the 16 and 17 December, but it also increases the overestimation of the large amplitude event between the 10 and 13 December.

4.3.4. January 1995. Europe is the largest anthropogenic source of C_m during January 1995, and the biogenic sources account for 60% of the amplitude of the variations in C_m. On the 20 and the 23 January, the amplitude of the modelled variations is too large. Since all anthropogenic source functions produce too large amplitude variations on these dates, the explanation for the discrepancy is likely to be found in the meteorological data and the model treatment of these. During the last days of January 1995 the amplitude of C_m is too low. The trajectories for this period indicate Arctic or eastern Siberian air as during the entire month (Fig. 2). Possible explanations for the small modelled amplitude are influence of anthropogenic or biogenic sources outside the model boundaries or too low biogenic emissions in the CCM3 estimate in Siberia.

4.3.5. Condensation nuclei and CO_2. In EH99, the correlation between the aerosol particle scattering coefficient, σ_{sp}, and C_m at Zeppelinfjellet is used as an indication of negligible biogenic sources for C_o in February 1993 and December 1994. To investigate the relations between C_o and "aerosol concentration" and anthropogenic and biogenic C_m we have chosen to study February 1993 in more detail. Due to lack of data on σ_{sp}, the number concentration of condensation nuclei, N_{cn}, is used instead. While σ_{sp} is a measure of the total mass of the aerosol particles, N_{cn} measures the number of aerosol particles. Both σ_{sp} and N_{cn} are indicators of anthropogenic sources whereas biogenic sources will have little influence on these quantities at least during winter.

In Fig. 4, C_m for February 1993 from anthropogenic and biogenic emissions is shown separately, and can be compared to C_o and N_{cn}. We find that the variations in C_m due to biogenic and anthropogenic emissions are well correlated. The source regions and source strengths for these two source types are similar. Due to the absence of major local sources, the Arctic CO_2 concentration is primarily determined by the transport to this region of CO_2 emitted elsewhere. This explains the covariation between the CO_2 of anthropogenic and biogenic origin at Zeppelinfjellet. Due to this covariation between CO_2 of anthropogenic and biogenic origin, the correlation between anthropogenic C_m variations and σ_{sp} or N_{cn} can not be used to exclude biogenic sources of the variations in C_o.

5. Conclusions

The synoptic variations of CO_2 during four winter months at an Arctic monitoring station have been studied to determine the sources of the observed CO_2 concentration. We find that the biogenic emissions of CO_2 effect the Arctic CO_2 concentration also during winter. This is in agreement with the results of Yuen et al. (1996) who find that biogenic emissions explain about 60% of a measured peak in the CO_2 concentration at Alert.

We also find the dominating anthropogenic source to be Europe, regardless of transport pathway to the station, and find no indications of the lower Ob-region as an area of large impact on Arctic CO_2 concentrations. This is in contradiction to the results obtained by Engardt and Holmén (1999), who only studied two winter months. Also, the source function used in their study is not appropriate to describe anthropogenic CO_2 emissions.

The few direct measurements of the biogenic fluxes of CO_2 (Dörr and Münnich, 1987; Zimov et al., 1993, 1996) indicate that the fluxes used in this study are a factor of 3–10 lower than the measured fluxes. However, the high direct flux numbers would, if taken as representative for larger regions, be incompatible with our results. The sum of the anthropogenic and biogenic emissions used in this study produce an amplitude of the same order as the observed amplitude at Zeppelinfjellet. The apparent contradiction

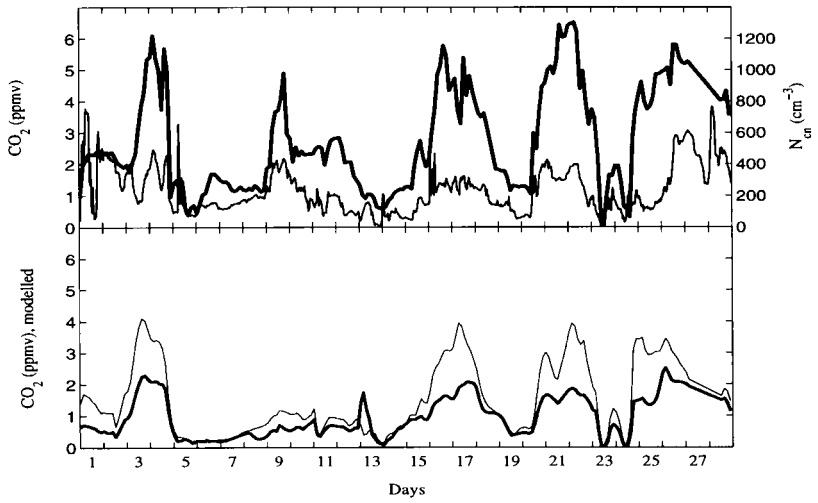

Fig. 4. Observations and model results for February 1993. In the top panel the observed CO_2 concentration(thick line) and the number concentration of condensation nuclei (thin line) are shown. The background variation of the observed concentration has been subtracted from the observed concentration. The curves of observed concentration are parallel shifted to obtain non-negative values of the concentration anomaly. In the bottom panel the modelled concentration using anthropogenic emissions (thick line) and biogenic emissions (thin line) is shown.

between local flux measurements and regional fluxes of CO_2 remains to be resolved.

So far, sources of Arctic CO_2 have been studied with the model for four winter months. Correlations of observed versus modelled concentrations are significant (at the 95% confidence level) for these 4 months together. Further, the amplitude of the modelled CO_2 variations is similar to the observed variations and for most of the time the timing is remarkably good. This shows that the model, used together with the anthropogenic and biogenic source functions, is a useful tool for investigating transport of CO_2 into the Arctic. To obtain further knowledge on long-range transport of polluted air to the Arctic region, we suggest the following. First, due to the uncertainties in the biogenic wintertime fluxes, more measurements of these fluxes are called for. This model study should be extended to comparisons with all available CO_2 and ^{14}C monitoring stations within the region. Also, to attempt to discriminate between biogenic and anthropogenic sources of the observed CO_2 at Zeppelinfjellet, ^{14}C sampling during CO_2 events could be useful. A study of the effect of the coarse vertical and horizontal resolution on modelled amplitudes should also be undertaken.

6. Acknowledgments

We thank M. Engardt for help with the model and for good advise. We also thank H. Rodhe for a critical review of the manuscript and two anonymous reviewers for constructive comments. The atmospheric monitoring activities performed by Stockholm University at Zeppelinfjellet are financed by the Swedish Environmental Protection Agency. This work was supported by the European Commission (through the ESCOBA project).

REFERENCES

Andres, R. J., Marland, G., Fung, I. and Matthews, E. 1996. A $1^\circ \times 1^\circ$ distribution of carbon dioxide emissions from fossil fuel consumption and cement manufacture, 1950-1990. *Global Biogeochemical Cycles* **10**, 419–429.

Benkovitz, C. M., Scholtz, M. T., Pacyna, J., Tarrasón, L., Dignon, J., Voldner, E. C., Spiro, P. A., Logan, J. A. and Graedel, T. E. 1996. Global gridded inventories of anthropogenic emissions of sulphur and nitrogen. *J. Geophys. Res.* **101D**, 29239–29253.

Bonan, G. B. 1996. *A land surface model (LSM version 1.0) for ecological,hydrological, and atmospheric studies. Technical description and user's guide.* NCAR Technical Note. National Center for Atmospheric Research, Boulder, Colorado, USA. NCAR/TN-417+STR.

Dörr, H. and Münnich, K. O. 1987. Annual variation in soil respiration in selected areas of the temperate zone. *Tellus* **39B**, 114–121.

Engardt, M., Holmén, K. and Heintzenberg J. 1996. Short-term variations in atmospheric CO_2 at Ny-Ålesund, Spitsbergen, during summer and winter. *Tellus* **48B**, 2881–2894.

Engardt, M. and Holmén, K. 1999. Model simulations of anthropogenic-CO_2 transport to an Arctic monitoring station during winter. *Tellus* **51B**, 194–209.

Fung, I. Y., Tucker, C. J. and Prentice K. C. 1987. Application of advanced very high resolution radiometer vegetation index to study atmosphere–biosphere exchange of CO_2. *J. Geophys. Res.* **92**, 2999–3015.

Hanna, S. R. 1994. Mesoscale meteorological model evaluation techniques with emphasis on needs of air quality models. In: *Mesoscale modeling of the atmosphere* (eds. R. A. Pielke and R. P. Pearce). American Meteorological Society, Boston, Massachusetts, USA, pp. 47–62.

Higushi, K. and Daggupaty, S. M. 1985. On variability of atmospheric CO_2 at station Alert. *Atmospheric Environment* **19**, 2039–2044.

Holmén, K., Engardt, M. and Odh, S.-Å. 1995. *The carbon dioxide measurement program at the Department of Meteorology at Stockholm University.* Report CM-84, Dept. of Meteorology, Stockholm University, Stockholm, Sweden, 38 pp.

McGrath, R. 1989. *Trajectory models and their use in the Irish Meteorological Service.* Memorandum no. 112/89. Irish Meteorological Service, Dublin, Ireland.

Olivier, J. G. J, Bouwman, A. F., Van der Maas, C. W. M., Berdowski, J. J. M., Veldt, C., Bloos, J. P. J., Visschedijk, A. J. H., Zandveld, P. Y. J. and Haverlag, J. L. 1996. *Description of EDGAR version 2.0. A set of emission inventories of greenhouse gases and ozone-depleting substances for all anthropogenic and most natural sources on a per country basis and on 1° × 1° grid.* RIVM/TNO report nr. 771060 002, RIVM, Bilthoven, The Netherlands.

Peterson, J. T., Komhyr, W. D., Harris, T. B. and Waterman, L. S. 1982. Atmospheric carbon dioxide measurements at Barrow, Alaska, 1973–1979. *Tellus* **34**, 166–175.

Robertson, L., Langner, J. and Engardt, M. 1999. An Eulerian limited area atmospheric transport model. *J. Appl. Met.* **38**, 190–210.

Schmidt, M., Graul, R., Sartorius, H. and Levin, I. 1996. Carbon dioxide and methane in continental Europe: a climatology, and ^{222}Radon-based emission estimates. *Tellus* **48B**, 457–473.

Wong, C. S., Chan, Y.-H., Page, J. S., Bellafay, R. D. and Petit, K. 1984. Trends of atmospheric CO_2 over Canadian background stations at Ocean Weather P, Sable Island and Alert. *J. Geophys. Res.* **89**, 9527–9539.

Yuen, C. W., Higushi, K., Trivett, N. B. A. and Cho, H. R. 1996. A simulation of a large positive CO_2 anomaly over the Canadian Arctic archipelago. *J. Met. Soc. Japan* **74**, 781–795.

Zimov, S. A., Semiletov, I. P., Davidov, S. P., Voropaev, Yu. V., Prosyannikov, S. F., Wong, C. S. and Chan, Y.-H. 1993. Wintertime CO_2 emissions from soils of northwestern Siberia. *Arctic* **3**, 197–204.

Zimov, S. A., Davidov, S. P., Voropaev, Yu. V., Prosisiannikov, S. F., Semiletov, I. P., Chapin, M. C. and Chapin, F. S. 1996. Siberian CO_2 efflux in winter as a CO_2 source and cause of seasonality in atmospheric CO_2. *Climate Change* **33**, 111–120.

Zondervan, A. and Meijer, H. A. J. 1996. Isotopic characterisation of CO_2 sources during regional pollution events using isotopic and radiocarbon analysis. *Tellus* **48B**, 601–612.