Atmospheric transport of carbon dioxide to a baseline monitoring station in northern Finland

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ABSTRACT

Interannual and seasonal variations in atmospheric transport to a baseline monitoring station at Pallas (67°58'N, 24°07'E) in northern Finland were examined. The transport was analysed through cluster analysis of three-dimensional 5-d backtrajectories during the period 1997–2003. The trajectory climatology shows that air mass advection from the north is most frequent-mostly at high wind speeds across the Arctic Basin and from northern Siberia-but during summer more stagnant flows from the Norwegian Sea are common as well. Western and Central Europe were found to be the second most important regions of influence for air arriving at Pallas, followed by atmospheric transport from west Russia and the Atlantic, respectively. The trajectory clusters were combined with measurements of carbon dioxide (CO₂) in order to examine the linkage between atmospheric large-scale circulation and CO2 concentration at Pallas. The Atlantic and Arctic air masses were associated with relatively small annual CO2 amplitudes at Pallas. In contrast, large concentration differences between the summer minimum and winter maximum were observed during periods of continental air mass transport from the south and the east. In particular the air masses originating from west Russia were associated with very low CO₂ concentrations during summer, indicating high photosynthetic activity of the terrestrial biosphere in this region. We analysed how the vertical motion of the trajectories affects the observed CO₂ at Pallas. The largest difference in CO₂ concentration between air parcels moving at low and high altitudes, respectively, was found during air mass advection from Europe and west Russia. This was especially true during the winter months when large CO₂ emissions in these areas, i.e. from fossil fuel combustion and the decomposition and respiration of the vegetation, in combination with stable stratification can give rise to very high CO₂ concentrations in air parcels transported close to the surface. The CO₂ time-series from Pallas was compared with CO₂ measurements made at the Mount Zeppelin station on Svalbard, illustrating the different characteristics—boreal and maritime, respectively—of the regions affecting the two monitoring sites.

1. Introduction

Today the carbon dioxide (CO_2) concentration in the atmosphere is increasing, largely due to emissions from the burning of fossil fuel. Because CO_2 is a greenhouse gas this increase is expected to lead to significant global climatic change during the coming decades (IPCC, 2001). In order to understand how the global carbon cycle will respond to such perturbations a good understanding of the present CO_2 fluxes to and from the atmosphere is required (Ciais et al., 1995; Rayner et al., 1999). An essential research tool for detecting, attributing and predicting these fluxes is long-term monitoring (cf. Enting and Mansbridge, 1989; Keeling et al., 1989; Tans et al., 1989; Ciais et al., 1995). For example, the multidecadal records of atmospheric CO_2 concen-

*Corresponding author. e-mail: tuula.aalto@fmi.fi trations from baseline observing stations at Mauna Loa, Cape Grim and elsewhere play an important role in the science of the carbon cycle and the Earth system (e.g. Keeling and Whorf, 2000).

In the present study a time-series of atmospheric CO₂ from a Global Atmospheric Watch (GAW) monitoring station at Pallas in northern Finland was analysed. The station is situated at the northernmost limit of the northern boreal forest zone in the sub-Arctic region. Here, as in the Arctic region, the atmospheric CO₂ concentration is highly influenced by long-range transport from mid-latitude source regions (cf. Conway et al., 1993; Engardt and Holmén, 1999; Brandefelt and Holmén, 2001; Aalto et al., 2002, 2003; Eneroth et al., 2003). The synoptic circulation to Pallas was examined through cluster analysis of 5-d back trajectories during the period 1997–2003. The established trajectory climatology was used to examine patterns of air mass advection and how these vary annually and seasonally. It is essential to monitor such

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variations in transport, since shifts in air mass statistics at a monitoring site can otherwise erroneously be interpreted as a trend in sources and sinks. For example, Higuchi et al. (2002) demonstrated that a significant portion of the observed quasi-decadal variation in the amplitude of the seasonal CO_2 cycle (an indicator of the metabolism of the terrestrial biosphere) at high-latitude stations in the Northern Hemisphere can mostly be accounted for by changes in the atmospheric circulation alone. In a similar way, Murayama et al. (2004) showed that a recognizable portion of the interannual variation in the growth rate of atmospheric CO_2 , particularly in mid- to high-latitude locations in the Northern Hemisphere, can be accounted for by changes in the atmospheric circulation.

To study the connection between CO_2 concentration and different transport situations the calculated trajectory climatology was applied to the Pallas CO_2 record. Previous studies have indicated that the Arctic is a major source region for air arriving at Pallas (Rummukainen et al., 1996; Aalto et al., 2002, 2003). Special attention was thus given to periods of air mass advection from the Arctic, for which the CO_2 observations at Pallas were compared with CO_2 measurements performed at the Mount Zeppelin station on Svalbard.

2. Methods

2.1. Monitoring of carbon dioxide

The Finnish Meteorological Institute (FMI) operates four separate stations at Pallas in northern Finland, measuring tropospheric air composition and related meteorological parameters. The stations are within 12 km of each other, and since 1994, together with FMI's Arctic Research Centre in Sodankylä, have been part of the Global Atmospheric Watch (GAW) programme. The area where the stations are located consists of a mixture of pine, spruce and birch forest and has no significant local or regional pollution sources. The main station at Pallas is located in the Pallas-Ounastunturi National Park on the top of Sammaltunturi hill $(67^{\circ}58'N, 24^{\circ}07'E)$ at 565 m above sea level (a.s.l.). The vegetation on the hill is sparse with the tree line about 100 m below the station. Sammaltunturi station is rarely within the surface inversion, making it an appropriate site for global background measurements. Further description of the CO₂ measurements and atmospheric research activities at Sammaltunturi station is given by Aalto et al. (2002) and Hatakka et al. (2003). In the present study CO₂ measurements from Sammaltunturi during 1998-2003 were analysed.

CO₂ time-series from the GAW monitoring station in Ny-Ålesund on Svalbard were used for comparison. The station is located on the Mount Zeppelin ridge (78°58′N, 11°53′E) at 474 m a.s.l. CO₂ measurements have been made at Mount Zeppelin station since 1989 and are described in detail by Holmén (1995) and Holmén et al. (1995).

2.2. Trajectories and classification

Three-dimensional 5-d back-trajectories arriving at Pallas at 925 hPa were calculated with the kinematic model FLEXTRA (cf. Stohl and Koffi, 1998; Stohl et al., 1999). This model utilizes meteorological data from the European Centre for Medium Range Weather Forecasts (ECMWF). The calculations were performed twice daily (at 00:00 and 12:00 UTC) for the period 1997–2003, resulting in a data set of 5112 trajectories. The trajectories were classified using cluster analysis, following Eneroth et al. (2003). Cluster analysis denotes a variety of multivariate techniques used to group similar objects together, whereby differences between individual elements within a cluster are minimized but differences between clusters are maximized Romesburg (1984). This method has the advantage over sector analysis in that in addition to direction it also takes wind speed into account. Wind speed is an important parameter, as it affects the time an air parcel spends over source/sink areas and it also gives an indication of the weather situation, e.g. low wind speeds imply stagnant weather conditions. In the present study Ward's minimum variance technique was applied, provided by the Matlab® standard statistical package. In the clustering procedure the latitude and longitude coordinates at 1-h intervals along the trajectories were used to calculate the distances between the trajectories. Each trajectory was initially defined as a separate cluster with zero spatial variance. The spatial variance is the sum of the squared distances between the latitude and longitude points of the cluster's component trajectories. The smaller the spatial variance, the more similar are the trajectories. Successive steps through the clustering process combine the two clusters that result in the minimum increase in total spatial variance (TSV), where TSV is the sum of all the cluster spatial variances. In the first steps of the clustering process the TSV increases greatly. In the succeeding steps it typically increases at a small rate. However, at some point the TSV again starts to increase rapidly. This "breaking point" indicates that the clusters being combined are not very similar and that the clustering should be stopped. The criterion defined by Stunder (1996) was used to decide when to stop the clustering. The criterion is met when the number of resulting clusters is more than three and less than 10, and the per cent increase in TSV at the break point is at least 30%.

3. Results

3.1. Climatology

Figure 1 shows the obtained climatology for trajectories arriving at Pallas at 925 hPa during 1997–2003. The station is affected by northerly transport at high wind speeds from the Arctic Basin and northern Siberia for 37% of the year (clusters 3 and 5) and at low wind speeds from the Norwegian Sea for 13% of the year (cluster 4). The second most important source region for air

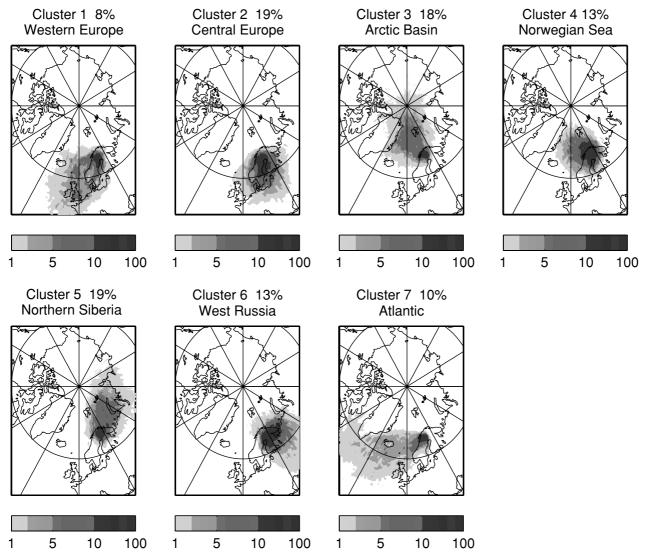


Fig 1. Density plots of trajectories arriving at Sammaltunturi at 925 hPa during 1997–2003. The different grey shadings enclose all grid squares containing values greater than 1, 2, 5 and 10% of the total number of trajectories. The percentage frequency of occurrence of each trajectory cluster is indicated in the figure. The average trajectory lengths in the different clusters are: cluster 1, 3990 km; cluster 2, 3050 km; cluster 3, 3840 km; cluster 4, 2890 km; cluster 5, 3600 km; cluster 6, 3100 km, cluster 7, 5410 km.

arriving at Pallas is Europe, with flow patterns from western and Central Europe (clusters 1 and 2) containing 27% of the trajectories. Thirteen per cent of the trajectories describe southeasterly transport from west Russia (cluster 6) and 10% of the trajectories denote maritime air masses that have passed over the Atlantic (cluster 7). The highest wind speeds, i.e. the longest 5-d trajectories, are observed during periods of westerly air transport from the Atlantic (cluster 7), but also during periods of southwesterly transport over western Europe (cluster 1) and northerly transport across the Arctic Basin (cluster 3). During periods of transport from Central Europe, the Norwegian Sea and west Russia (clusters 2, 4 and 6) low wind speeds and short 5-d trajectories are observed at Sammaltunturi.

Table 1 shows the interannual variation of the distribution of trajectories between the transport clusters. The prevalence of the different flow patterns differs from year to year. In particular, 1998 differs from the other years. During this year the transport was displaced eastwards favouring trajectories originating from northern Siberia and west Russia (clusters 5 and 6) at the expense of air mass transport from western Europe, the Norwegian Sea and the Atlantic (clusters 1, 4 and 7). The same transport deviation during 1998 was observed by Eneroth et al. (2003), who examined trajectories arriving at Ny-Ålesund (78°58′N,11°53′E), Svalbard during 1992–2001.

There is also a seasonal variation in the atmospheric transport to Pallas. Table 2 shows the mean percentage distribution of

Cluster	1997	1998	1999	2000	2001	2002	2003	Mean	SD	
1	7.9	4.4	9.6	11.3	6.8	7.9	7.5	7.9	2.2	
2	18.6	20.3	22.2	17.9	17.7	16.6	21.9	19.3	2.2	
3	21.0	15.1	15.3	16.8	19.5	15.5	20.4	17.6	2.6	
4	14.4	7.7	9.3	14.3	11.1	18.1	13.0	12.6	3.5	
5	18.1	27.1	17.3	14.8	19.3	20.7	18.5	19.4	3.8	

14.5

11.1

8.4

12.9

9.5

9.2

13.2

9.9

4.0

2.2

15.8

9.0

Table 1. Percentage of trajectories in each cluster during different years. The two right-hand columns show the mean distribution and standard deviation during 1997–2003

Table 2. Percentage of trajectories in each cluster during different months in 1997–2003

14.7

11.6

19.5

6.0

Cluster	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
1	5.2	6.9	4.4	7.4	7.4	10.8	15.5	10.6	9.4	7.6	7.4	7.4
2	6.5	7.0	7.8	7.5	6.7	6.5	10.2	7.3	11.1	8.8	11.1	9.4
3	12.0	6.5	8.2	12.4	8.2	6.4	4.0	6.0	9.2	9.0	7.0	11.0
4	6.7	3.6	5.0	6.1	12.3	15.1	14.3	12.5	7.8	6.4	4.0	6.2
5	9.2	7.7	11.9	9.1	11.7	8.2	4.4	7.3	4.6	10.1	7.1	8.8
6	6.2	12.1	8.1	7.5	4.6	6.5	8.1	9.2	8.1	9.2	14.3	6.1
7	12.8	11.2	11.8	4.7	7.5	6.3	8.5	9.9	7.5	6.3	4.7	8.7

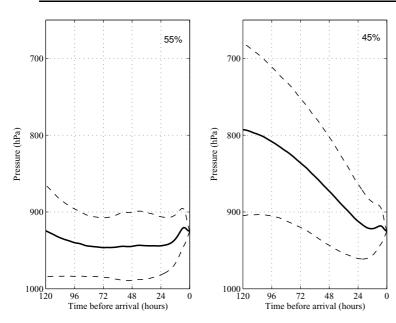


Fig 2. The vertical position of low and high trajectories arriving at Sammaltunturi at 925 hPa during 1997–2003. The solid line denotes the mean altitude of the trajectories and the dashed lines denote the standard deviation. The numbers show the percentage distribution of trajectories between the two heights.

trajectories in each cluster for each month. The transport clusters describing transport from western Europe (cluster 1) and the Norwegian Sea (cluster 4) are most frequent during summer. On the other hand, northerly transport across the Arctic Basin and from northern Siberia (clusters 3 and 5) occurs rarely during summer. The easterly (cluster 6) and the westerly (cluster 7) air masses from west Russia and the Atlantic, respectively, are most frequent during autumn and winter.

It is important to take into account the vertical level of the trajectories when interpreting time-series of atmospheric species. This is because the air is greatly affected by surface fluxes during its residence time in the planetary boundary layer whereas it is cut off from direct exchange with sources and sinks at the ground when transported in the free troposphere. The trajectories in the climatology were classified as either low or high using the pressure along their paths as a cluster variable. Fifty-five per cent of the trajectories were categorized as low and 45% as high (see Fig. 2). The percentage distribution between the two heights for each trajectory cluster is shown in Table 3. Northerly (clusters 3, 4 and 5) and westerly (cluster 7) transport patterns have

6

7

10.4

9.6

Table 3. Percentage of low and high trajectories in each cluster during 1997–2003

	Cl. 1	C1. 2	Cl. 3	Cl. 4	Cl. 5	Cl. 6	Cl. 7
High	40	41	47	52	48	40	49
Low	60	59	53	48	52	60	51

Table 4. Percentage of low and high trajectories in each month during 1997–2003

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
High	49	45	48	46	41	41	40	39	52	47	48	51
Low	51	55	52	54	59	59	60	61	48	53	52	49

the highest percentage of trajectories advected at high altitudes. Most often the air from these areas is colder than the air over Finland, thus the air descends as it approaches Pallas (cold air advection). The percentage distribution of low and high trajectories between the months was also examined and is presented in Table 4. It can be seen that high (low) trajectories are most frequent during winter (summer). This can be explained in the same way as the distribution of high and low trajectories between the different clusters. Descending trajectories at Pallas are more frequent during winter due to the sinking of cold air.

3.2. The influence of transport on carbon dioxide at Pallas

The climatology was applied to the Pallas CO₂ data in order to examine the influence of different atmospheric flow patterns on

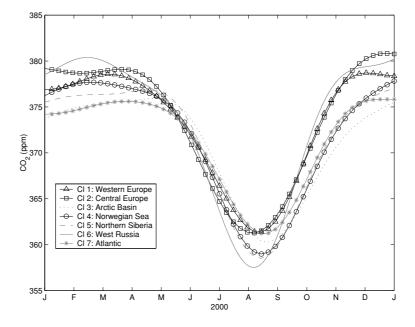
the CO_2 concentration. The data were divided into seven separate groups according to the trajectory climatology. The mean value of the CO_2 hourly means between ± 1 h of the arrival time of the trajectory was calculated and used in the analysis. In order to estimate the long-term growth trend and the smooth annual variation a harmonic function was fitted to the data. The following fitting algorithm was used (cf. Kahaner et al., 1989; Tans et al., 1989):

$$x(t) = x_0 + x_1 t + \sum_{k} \left[a_k \sin\left(\frac{z\pi tk}{T}\right) + b_k \cos\left(\frac{2\pi tk}{T}\right) \right]$$
(1)

where x is the CO₂ concentration and t is time (if expressed in years then sequence T equals 1). The coefficients to be fitted include constant x_0 , growth trend x_1 and coefficients for the harmonic function, a_k and b_k , where k = 1 to 3.

Figure 3 shows the harmonic functions fitted to the seven data records. It can be seen that the maritime air masses are associated with a smaller amplitude of the annual CO₂ cycle compared with the continental air masses. The large difference in concentration between the summer minimum and the winter maximum in connection with continental air mass transport is primarily caused by the yearly cycle of photosynthesis and respiration of the terrestrial biosphere, but is also due to anthropogenic wintertime emissions of CO₂ over Europe and Russia (cf. Engardt and Holmén, 1999; Brandefelt and Holmén, 2001). The fitted curves of the data records associated with southerly transport from western and Central Europe (clusters 1 and 2) are doublepeaked, with a small maximum in autumn/early winter and a larger one later in the winter. This was also observed in a previous study by Aalto et al. (2003), classifying CO₂ measurements at Sammaltunturi according to sector analysis of the trajectories. There is also the weak autumn/early winter maximum in

Fig 3. Harmonic functions fitted to CO₂ observations at Sammaltunturi during 2000 for each of the seven trajectory clusters. The amplitudes of the annual cycle of the CO₂ concentration for each cluster are: cluster 1, 17 ppm; cluster 2, 20 ppm; cluster 3, 16 ppm; cluster 4, 19 ppm; cluster 5, 19 ppm; cluster 6, 23 ppm, cluster 7, 15 ppm.



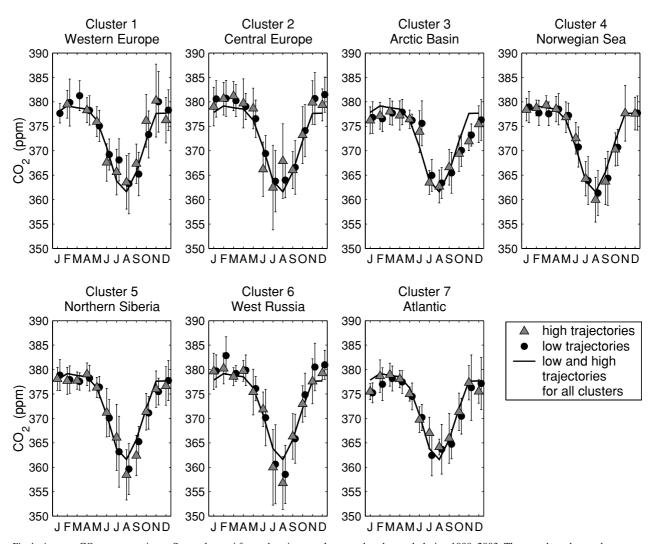


Fig 4. Average CO₂ concentration at Sammaltunturi for each trajectory cluster and each month during 1998–2003. The error bars denote the standard deviations of the mean values. The grey triangles and black circles denote high and low trajectories, respectively. The curve shows the average CO₂ concentration for all data, regardless of transport pattern.

connection with transport from west Russia (cluster 6) and the Atlantic (cluster 7).

Figure 4 shows monthly means of CO₂ concentration for the seven clusters for both low and high trajectories. The trajectories describing transport from west Russia (cluster 6) and from western and Central Europe (clusters 1 and 2) are associated with a higher than average CO₂ concentration during winter, reflecting long residence times over regions with high wintertime CO₂ emissions from the vegetation and anthropogenic sources. The influence of the terrestrial biosphere is especially noticeable in air masses from west Russia (cluster 6) with very low CO₂ concentrations during summer, indicating high photosynthetic activity in this region. During northerly and westerly transports lower than average CO₂ concentrations are observed during autumn and winter. This is due to the much smaller CO₂ emissions from

fossil fuel combustion and terrestrial decomposition and respiration in the Arctic region and over the Atlantic compared with Europe and Russia. The trajectories from western and Central Europe as well as west Russia, i.e. air masses transported over areas which are sources of anthropogenic CO_2 , are associated with high CO_2 variability, which is reflected in Fig. 4 as large standard deviations of the monthly concentrations. These air masses also show the largest differences in CO_2 concentration, depending on the vertical level of the trajectories. The trajectories advected at low altitudes are most often associated with higher CO_2 concentration compared with the more elevated trajectories. This is especially true during the winter months when the combination of stable atmospheric stratification and large anthropogenic and biogenic surface emissions can give rise to very high CO_2 concentrations in air parcels transported close to the surface.

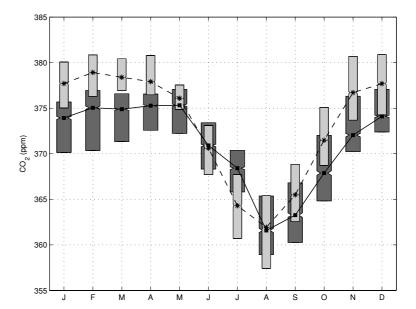


Fig 5. Box diagrams of CO_2 concentration at Sammaltunturi and Mount Zeppelin during 1998–2003. The boxes have borders at the lower and upper quartile values. The stars and the dashed line denote median CO_2 concentration at Sammaltunturi station for each month. The squares and the solid line denote median CO_2 concentration at Mount Zeppelin station for each month.

3.3. Carbon dioxide on Svalbard and in northern Finland

Previous studies (cf. Rummukainen et al., 1996; Aalto et al., 2002, 2003) have shown that the Pallas region is frequently affected by Arctic air masses. We therefore compared the CO2 time-series from Sammaltunturi station with CO2 measurements made at Mount Zeppelin station on Svalbard. Figure 5 shows calculated monthly means of CO2 concentration at Sammaltunturi and Mount Zeppelin during 1998-2003. The annual CO₂ amplitude is higher at Sammaltunturi station (18 ppm) than at Mount Zeppelin station (15 ppm). This reflects the comparatively greater influence of the terrestrial vegetation and anthropogenic emissions at Pallas. On average the observed CO₂ concentration is about 2 ppm higher at Sammaltunturi than at Mount Zeppelin, with a maximum and minimum difference in winter and summer, respectively. As a result of the photosynthetic activity of the terrestrial biosphere in summer, lower average CO₂ concentrations are observed at Sammaltunturi than at Mount Zeppelin in July. The two monitoring stations both participate in the NOAA Climate Monitoring and Diagnostics Laboratory (CMDL) Carbon Cycle Greenhouse Gases (CCGG) cooperative air sampling network, which collects flask air samples approximately weekly from a globally distributed network of sites. During the time period of the present study the flask samples from Mount Zeppelin show higher CO₂ concentrations (around 1.6 ppm) than the continuous measurements. There is no such discrepancy between the Sammaltunturi continuous data and the NOAA flask samples. The Mount Zeppelin deviation will be investigated further. The absolute figures for the CO₂ concentration differences in Figure 5 should thus be regarded with some caution.

To examine what effect the general circulation has on the relation between CO₂ at Sammaltunturi and Mount Zeppelin scatter diagrams of CO₂ concentrations were generated concen-

trations for each trajectory cluster. These diagrams, as well as calculated correlation coefficients, are presented in Fig. 6. As expected, the strongest correlation was found in connection with the northerly transport flows to Pallas, in particular during periods of air mass advection across the Arctic Basin (cluster 3). Furthermore, the difference in CO₂ concentration between the stations is about the same throughout the year in this cluster; approximately 1 ppm higher at Sammaltunturi than at Mount Zeppelin. In contrast there is a clear seasonal variance in the concentration differences in connection with air mass advection from Europe (clusters 1 and 2) and from west Russia (cluster 6). For these flow patterns the CO₂ concentration is markedly higher at Sammaltunturi than at Mount Zeppelin during winter, whereas it often happens that a lower CO₂ concentration is observed at Pallas than on Svalbard during summer. These concentration gradients between northern Finland and Svalbard should be further analysed, for example through atmospheric transport model calculations. Transport models (Chevillard et al., 2002; Kjellström et al., 2002) allow for mixing and quantitative experiments with source and sink distributions that can be used to test the interpretation of the trajectories for consistency.

4. Summary and conclusions

Cluster analysis was applied to a 7-yr trajectory database to establish an air mass transport climatology for Pallas. Seven major transport routes to Pallas were identified. The distribution of trajectories between these classes varies with season and from year to year. However, some general features apply to all years during the 7-yr period. The climatology shows that air mass advection from the north occur most frequently—mostly at high wind speeds across the Arctic Basin and from northern Siberia—but during summer more stagnant northerly flows from the Norwegian Sea are common as well. Western and Central Europe were

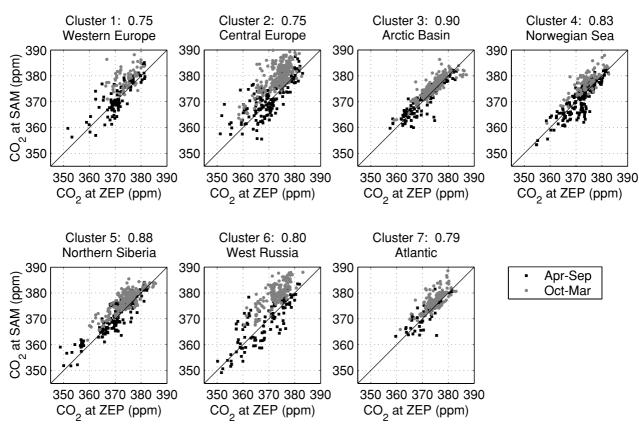


Fig 6. Scatter diagrams of CO_2 concentration at Sammaltunturi and Mount Zeppelin for each cluster. The grey dots and black squares denote CO_2 concentrations observed during October to March and April to September, respectively. The diagonal lines indicate equal CO_2 concentrations at the two stations. Calculated correlation coefficients between the two CO_2 records for each cluster are indicated in the headings.

found to be the second most important regions of influence for air arriving at Pallas, followed by air mass advection from west Russia and the Atlantic respectively. This result is in accordance with previous studies by Rummukainen et al. (1996) and Aalto et al. (2002, 2003) studying atmospheric transport to Pallas with the means of trajectories. However, the observed year to year variance of the prevalence of the different transport patterns implies that in order to avoid misinterpretations of the observed atmospheric time-series at Pallas the air mass statistics of the area should be continuously surveyed.

The climatology was applied to the CO_2 measurements made at Pallas in order to examine the influence of different flow patterns on the variability of atmospheric CO_2 . The largest concentration difference between winter maximum and summer minimum was observed in connection with continental air mass transport from the south and the east. In particular, the air originating from west Russia was associated with very low summertime CO_2 concentrations, indicating that the Russian forest acts as a large terrestrial CO_2 sink during summer. The maritime Atlantic and Arctic air masses were associated with relatively small annual CO_2 amplitudes. This is due to the smaller influence of terrestrial biosphere fluxes and anthropogenic CO_2 emissions in these areas compared with Russia and Europe.

The vertical level of the trajectories was found to have an influence on the observed CO_2 concentration at Pallas, especially during air mass advection from Europe and west Russia in winter. This implies that careful consideration should be given not only to the two-dimensional transport but also to the vertical motion of air parcels when interpreting observed CO_2 timeseries.

The Sammaltunturi CO2 record was compared with CO2 measurements made at Mount Zeppelin station on Svalbard. Both the amplitude of the annual CO2 cycle and the annual-mean concentration were higher at Sammaltunturi than at Mount Zeppelin, reflecting the comparatively larger influence of CO₂ emissions from anthropogenic sources and the terrestrial biosphere at Pallas. The strongest correlation between the stations was found during periods of northerly transport to Pallas. The difference between the two CO₂ records was approximately the same throughout the year during these periods of Arctic air mass advection. In contrast there was a clear seasonal variation in the concentration difference between the stations in connection with atmospheric transport from Europe and from Russia. The comparison illustrates that the concentration gradient between Mount Zeppelin and Pallas is more or less pronounced depending on how direct the transport is between the stations. Climatic (and seasonal) shifts in which air is arriving at the individual stations must be elucidated in order to interpret this gradient.

The application of the trajectory climatology to CO_2 data in this study illustrates the importance of analysing transport patterns to monitoring stations, i.e. one must have an understanding of the seasonal and annual variations/trends in the atmospheric circulation before drawing any conclusions about changes in measured concentrations. The method used is generally applicable and may in future studies advantageously be applied to atmospheric components other than CO_2 measured at Pallas.

5. Acknowledgments

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