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Influence of air mass source sector on variations in CO_2 mixing ratio at a boreal site in northern Finland

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CO_2 mixing ratio in air masses coming from different source sectors was studied at Pallas measurement station in Lapland. Source sectors were defined using back trajectories and wind direction measurements. Air masses from the North and West sectors showed an annual variation of 17 ppm, possibly affected by a long range transported marine air. A larger variation of 20 ppm was observed in air masses from the more continental South and East sectors. During late autumn mixing ratios in air masses from the South sector were high in comparison with the other sectors. Different methods for a source sector definition were considered for the site, located in a contoured terrain. 52%–73% of wind direction-based source sector definitions agreed with trajectory-based definitions. However, the number of cases with reliable sector definitions may remain low when considering all observations. Different definition methods can cause differences of the order of 1 ppm in sectorially selected monthly mean CO_2 mixing ratios.

Introduction

Tropospheric CO_2 mixing ratios have been monitored since the late 1950s (Keeling *et al*. 1976). At present the measurement network has grown into global extent with versatile site locations. The long lifetime of CO_2 enables episode detection over long distances. In order to separate influences from different types of sources and sinks, it is therefore necessary to examine the air parcel dynamics related to stations together with land use information. A measured mixing ratio can be classified as local or non-local (e.g. Ramonet *et al*. 1996), and the source region for a long-range-transported air parcel can be estimated from backward trajectories or wind direction measurements (e.g. Haagenson and Sperry 1989, Bousquet *et al*. 1996, Engardt *et al*. 1996, Higuchi *et al*. 2003). Classification of the experimental results is required in many integrated databases (GLOBALVIEW-CO_2: Cooperative Atmospheric Data Integration Project — Carbon Dioxide. CD-ROM, NOAA CMDL, Boulder, Colorado (available on the internet at ftp.cmdl.noaa.gov, path: ccg/co2/GLOBALVIEW), AEROCARB: European Atmospheric CO_2 Database, LSCE, Gif-sur-Yvette, France (available on the internet at http://www.aerocarb.cnrs-gif.fr)).

Determination of the origin of air parcels using wind direction measurements is more difficult if the measurement site is located in a mountainous region. The wind field can be distorted due to surrounding mountains, especially if they are higher than the location of the site, which may lead to misinterpretation of the measured wind direction. The Pallas station (567 m above sea level) is located on top of a hill that belongs to a chain of other hills, the highest top reaching 800 m above sea level. The station is situated close to the northern limit of the boreal forest zone in a very sparsely-populated region. The location of the Pallas site is continental, but the Atlantic coastline at about 300-km distance is reachable by a few days of long range transport. In this article we try to clarify the rules by which CO_2 measurements at Pallas can be classified using wind direction measurements and trajectories. The possibility of separating marine *vs*. continental influences at the site will also be considered.

Materials and methods

Measurements were made in Pallas–Ounastunturi National Park, characterized locally and regionally by a very limited number of pollution sources. The nearest center of population, Muonio, with 2500 inhabitants, is located at 19-km distance from the measuring site. The nearest towns are Rovaniemi, 35 000 inhabitants, at about 170-km distance and Kiruna, 24 000 inhabitants, at about 160-km distance. CO_2 is measured at the top of Sammaltunturi hill (67°58´N, 24°07´E,), located about 300 m above surrounding terrain and 560 m above sea level. The top of the hill is treeless and the sparse vegetation consists mainly of mosses and lichens. The treeline is about 100 m below the station. The forest consists of mixed species, mainly Scots pine (*Pinus sylvestris*), Norway spruce (*Picea abies*) and downy birch (*Betula pubescens*). Wetland areas and lakes can also be found within a few kilometers distance.

Continuous CO_2 measurements were started at Pallas in October 1996 by the Finnish Meteorological Institute (FMI). CO_2 is measured as one-minute means with a LiCor nondispersive infrared (NDIR) gas analyzer using one reference gas and three calibration gases. These working standard mixing ratios are measured against WMO/CCL primary standards once every three months. The sample is collected through heated stainless steel tubing at about 7 m above ground (567 m above sea level). The CO_2 data were analysed in order to select well-mixed air parcels free from dominating local influences and instrumental errors. Measurements made during station visits and equipment testing were removed. To accept an hourly CO_2 mixing ratio, a mean wind speed was required to be higher than 2 m s^{-1}, standard deviation of hourly mean lower than 1 ppm and subsequent hourly values less than 1 ppm apart from each other. These limitations act to remove the local fluctuations while preserving the rather fast but monotone rise or decrease in hourly-mean mixing ratios during changes in large scale weather patterns. The selection procedure removed about 14% of the mixing ratios from the original data set. A number of other species and meteorological variables are also measured at the site. Measurements at a weather station situated at the top of Laukukero (68°04´N, 24°02´E, 765 m above sea level), within 11 km distance of Sammaltunturi, are also utilised in this work. A more detailed description of the measurement set-up is presented in Aalto *et al*. (2002).

Air parcel trajectories arriving at Pallas were utilized in the analysis. Three-dimensional trajectories were generated using a kinematic trajectory model FLEXTRA (e.g. Stohl *et al*. 1999, Stohl and Koffi 1998, Stohl and Wotawa 1995) which utilizes numerical meteorological data from a European Centre for Medium-Range Weather Forecasts (ECWMF) MARS database. Trajectories were calculated for June 2001–May 2002 once in every three hours starting at an arrival time of 00:00 UTC (= local standard time –2 h) at 925-hPa level. The path and height of a trajectory were calculated once every hour and five days backward in time. The five-day trajectories are long enough to extend from Pallas to the relatively unpolluted polar region or to the large source areas of CO_2 in Central Europe, thus providing a tool for studying

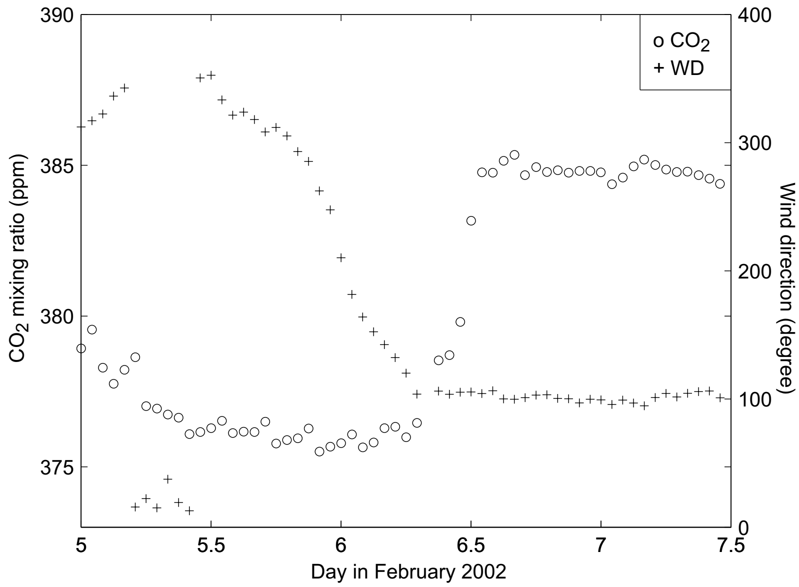

Fig. 1. Hourly means of CO_2 mixing ratio and wind direction observed at Pallas during February 2002. Wind direction is expressed as the direction where wind is coming from (degrees clockwise from north).

different types of natural and anthropogenic influences. It has to be remembered, though, that the atmospheric turnover time of CO_2 is several years and that the mixing ratio of CO_2 in air arriving at the site results from effects of numerous events (e.g. oceanic contact, biosphere contact, mixing and advection) along the trajectory. It is not straightforward to evaluate the relative importance of historic events to the observed mixing ratio. However, trajectories provide qualitative information by showing whether the air parcel has passed over a known source or sink area.

Results

A typical series of wintertime CO_2 observations at Pallas is shown in Fig. 1, where a relatively constant mixing ratio was followed by a rapid change coinciding with a change in wind direction. The Pallas site is affected by continental air masses crossing over boreal forests surrounding the site. However, marine influences are possible due to long range transport from the Atlantic Ocean and Baltic Sea. Furthermore, anthropogenic sources in the northern parts of Central Europe may have an effect on the CO_2 signal measured at Pallas (Aalto *et al*. 2002). The combination of these sources and sinks results in an average annual CO_2 cycle and growth rate shown in Hatakka *et al*. (2003). In this work the annual cycle is determined for different sets of CO_2 measurements classified according to wind direction and trajectories.

Source sectors indicated by the wind direction

The data set (hourly means) from October 1996 to October 2002 was divided into four separate records according to the wind direction sectors from where the air masses arrived (East: 45°–157°, South: 157°–240°, West: 240°–293°, North: 293°–45°, *see* Fig 2). The choice of sectors represents roughly the characteristics of the region: the West and North are marine sectors, while the East and South are more continental sectors. Extremely high CO_2 mixing ratios indicative of anthropogenic emissions are most often observed in air masses from the South, whereas the North sector usually corresponds to very clean air masses (*see* also Brandefelt and Holmen 2001). The wind directions were measured at the Sammaltunturi station.

Daily medians of the CO_2 mixing ratio were calculated separately for each wind sector (Fig. 3). If more than eight hourly means of CO_2 were assigned to a same wind sector during the

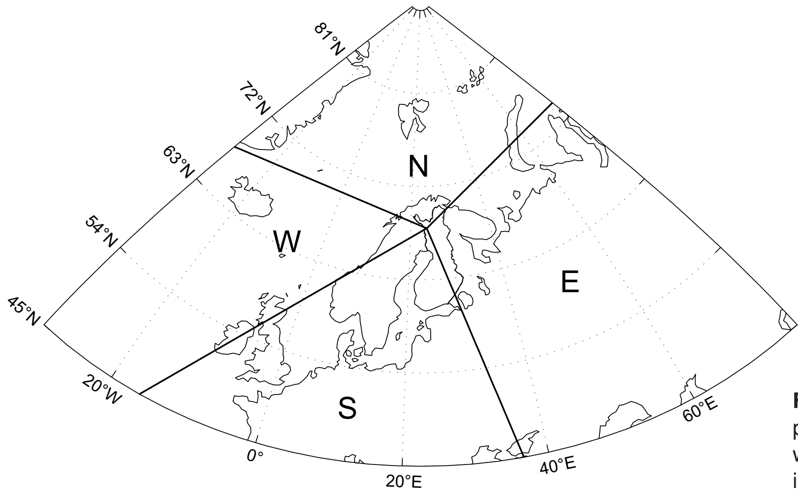

Fig. 2. Source sectors for air parcels observed at Pallas site, which is located at the point of intersection of the lines.

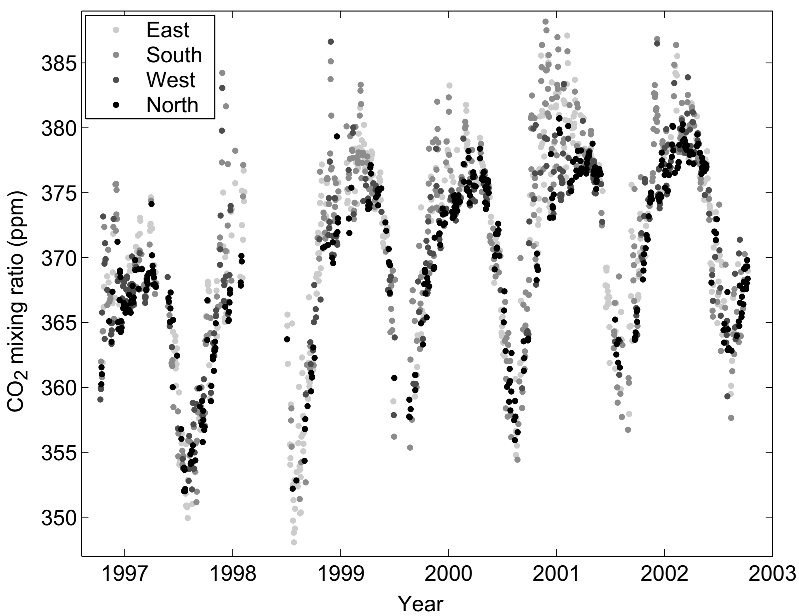

Fig. 3. Daily medians of CO_2 mixing ratio observed at Pallas during 1996–2003. Different shades of grey refer to the four source sectors for air parcels (Fig. 2) indicated by wind direction.

day, a median was calculated from those hourly means and added to the data set corresponding to the wind sector. The wind direction often stayed in one sector for more than one day and then changed during a few hours. The annual cycle was solved by fitting a harmonic function (six coefficients) with a first-degree polynomial to the daily values of the CO_2 mixing ratio (*see* Aalto *et al*. 2002). The phase of the annual cycle was rather similar for all the sectors (Fig. 4). Noticeable differences can be seen only in the annual cycle of the North sector, including a delayed mixing ratio increase during autumn and a delayed decrease during spring. This is supported by earlier observations, in which the annual cycle of some marine stations was delayed from the average annual cycle at Pallas (Aalto *et al*. 2002).

The North and West sectors showed smaller yearly variations (about 17 ppm) than the East and South sectors (about 20 ppm). This may indicate the activity by the terrestrial vegetation (the lowest summer values are observed in East), and also the effect of anthropogenic sources (*see*

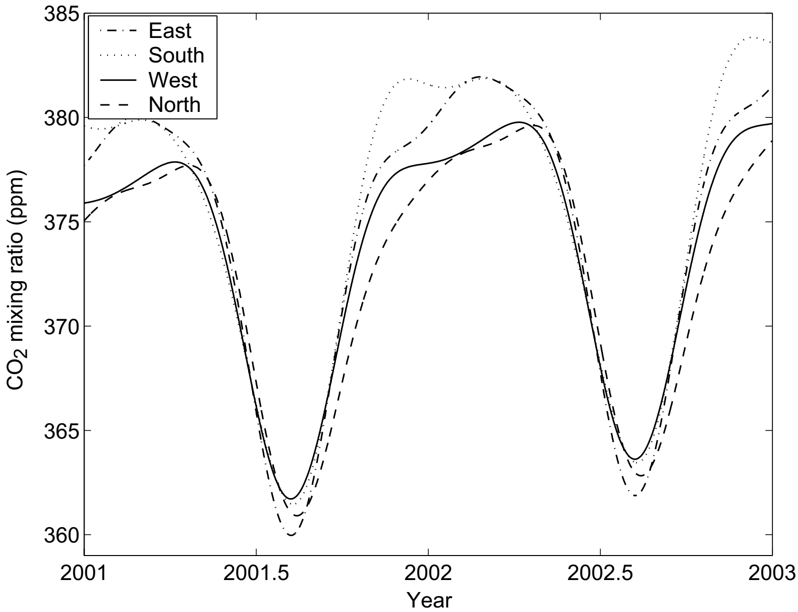

Fig. 4. Annual CO_2 cycles at Pallas. Curves are fitted to observed mixing ratios in air masses coming from different source sectors. Sectors are determined according to wind direction at Sammaltunturi.

also Brandefelt and Holmen 2001, Higuchi *et al.* 2003). The double-peaked shape of the South curve indicates high autumn-early winter values. A similar, but weaker, effect can be detected in the West curve. The early rise in the West curve actually results from only few very high daily values (Nov 1997, Nov 1998, Dec 2001). The trajectories related to those days indicate that the air masses actually passed over Central Europe before arriving at Pallas via the Atlantic Ocean. Those days should therefore be classified into the South sector, and the resulting shape of the West curve would be close to those in the other sectors. This example shows how important the trajectories can be in explaining the observed mixing ratios. The current hourly data set showed that the South sector contributed only 18%–19% of the air masses arriving to Pallas during spring and summer, while the respective percentages for autumn and winter were 37% and 26%. Therefore, during the late autumn and early winter, there is a greater possibility for observing exceptional episodes of elevated mixing ratios. The anthropogenic emissions also increase and get more dispersed towards winter, and the photosynthetic activity of terrestrial plants decreases rapidly while the net respiration may still continue at a considerable level. The autumn and winter episodes require further studies using simultaneous measurements of other species which may reveal the anthropogenic/biogenic origin of CO_2.

The other station in the Pallas region can be used to verify the sector determination. Wind direction at the Laukukero station is probably measured in a less distorted wind field because the station is situated at a 200-m higher altitude than the Sammaltunturi station. New CO_2 time series were created using only those observations in which the two stations indicated the same source sector (78% of the cases). The results were qualitatively similar, i.e. the North and West sectors had smaller amplitudes than the East and South sectors, and the winter maximum in the South sector was broader and started earlier than in other sectors (Fig. 5). Differences were found in the summer minimum of the South sector, which decreased about 1.5 ppm. The autumn peak in the West sector curve was also weaker. The selection procedure for reducing local influences was also relatively insensitive to changes. For example, the use of unselected data set resulted in less than a 0.4-ppm change in the yearly amplitude.

Wind direction *vs.* trajectories

Both wind direction and trajectories can be used to define a source region. Trajectories can be, for

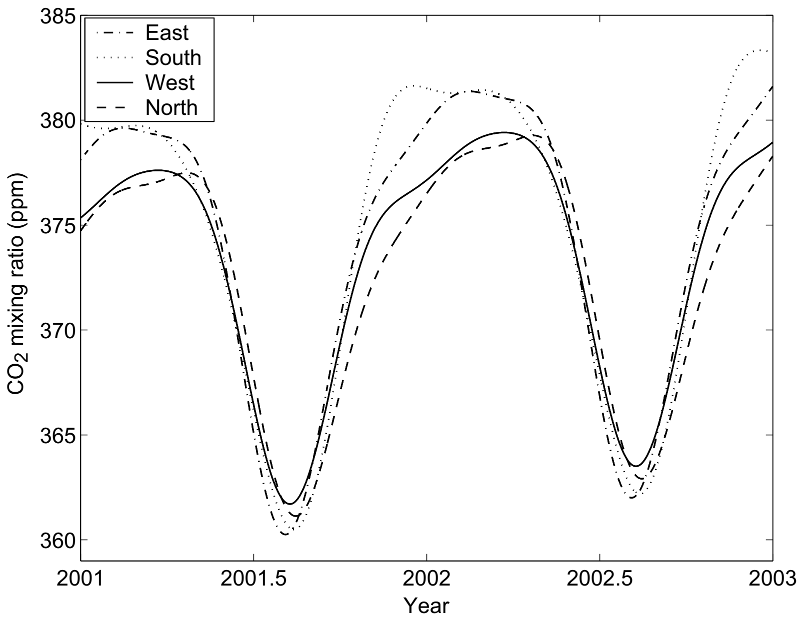

Fig. 5. Annual CO_2 cycles at Pallas. Curves are fitted to observed mixing ratios in air masses coming from different source sectors. Sectors are determined according to wind direction at Sammaltunturi and Laukukero.

example, used as in Aalto *et al*. (2002) where the source region was determined according to number of hours of travelling above a certain area (*see* also Rummukainen *et al*. 1996, Virkkula *et al*. 1998, Paatero and Hatakka 2000). The source region is not limited to certain sectors when using trajectories, for example the trajectories circulating in the vicinity of a site can be excluded from the investigation. However, in order to be consistent with the wind direction classification above, the trajectory data were first forced through a similar sector division. By doing this we were able to compare how well the wind direction actually corresponds to a certain sector.

The trajectory set from June 2001 to May 2002 was utilised (2779 trajectories). Three categories were defined for each trajectory to make it belong to a certain sector. If the trajectory did not match even the lowest category, it was rejected. The categories were based on the number of hours the trajectory spent in a certain sector. The highest limit was 90% of the total number of hours of a single trajectory in a certain sector (resulting number of trajectories approved was 782 out of 2779) and lowest limit 50%. Using these limits the number of cases can be calculated where the wind direction at the time of arrival of the trajectory and the trajectory classification indicate the same source sector. The percentages of the "right guess" cases from the number of approved trajectories are presented in Table 1. In order to improve the sector classification according to wind direction, an average of the wind direction over a few hours before arrival of the trajectory was used. In practice, if an average of three hours was used instead of a one-hour average, the percentages would be only 1%–2% higher. Essentially, the percentages in Table 1 indicate that if we have been able to identify the sector using trajectories, also the wind direction gives the same sector at least in 52% of the cases and at best in slightly over 70% of the cases. The classification according to wind direction was more often correct when the classification according to a trajectory was also more rigid. The general idea of equating the wind direction with the trajectory seems thus to be acceptable.

Table 1. Degree of agreement between trajectory and wind direction based sector classifications for classified trajectories.

Confidence level of sector classification according to trajectory	Percentage of successful sector classifications according to wind direction
90% (782 trajectories)	$567/782 \times 100 = 73\%$
75% (1362 trajectories)	$813/1362 \times 100 = 60\%$
50% (2556 trajectories)	$1330/2556 \times 100 = 52\%$

Low percentages were obtained when the results were converted to apply to all trajectories (Table 2). This means that half or less of all cases can be classified in a well-justified manner using wind direction measurements. Especially 90% and 75% levels showed very low numbers. On the other hand, the number of cases where the sector is definitely incorrectly identified (Table 2, third column) was rather small for the 90% and 75% levels.

In addition to the different sector determinations, about 3%–17% (corresponding to 90%–50% of the five-day trajectory path) of all trajectories circulated in the vicinity of the measurement site, i.e. inside a continental, relatively unpolluted region determined by 65°N, 71°N, 15°E and 29°E (region I in Aalto *et al.* 2002). These cases can not be detected using only wind direction measurements, and they should be excluded when discussing the transport from a distant source region.

Variations of monthly CO_2 mixing ratios according to source sector definition method

Monthly mean CO_2 mixing ratio is often utilized in global studies. If CO_2 values are to be selected according to a source sector, it is important to find out how much the resulting mean mixing ratios differ according to the source definition method. We focus here on monthly-mean mixing ratios during 2001–2002 because at present we do not have a consistent series of trajectories covering the six-year-long period of CO_2 measurements.

Averaging of CO_2 was performed for each source sector using wind direction measurements and, alternatively, trajectory crossings for the sector determination. Air masses observed at the site between trajectory arrivals were assumed to refer to the same source region as the latest trajectory, and an average mixing ratio over three hours, including the hour of the trajectory arrival, was utilized. Wind directions were also averaged over the same three-hour periods. Each three-hour-average mixing ratio was assigned to a source sector, as indicated by the trajectory which arrived at the site during the corresponding three-hour time window. The procedure was repeated using the wind direction for the source sector determination. Thus, two data sets were created where the same mixing ratios were distributed in a different way among sectors.

Monthly means were calculated from the three-hour-average mixing ratios for each sector using the two data sets. The maximum number of monthly means per data set is 48 (number of sectors times number of months), but only 22 were used because of low number of data points during the month. Each source sector was now connected with two separate monthly-average mixing ratios as indicated by wind directions and trajectories. These monthly means are plotted against each other in Fig. 6. The standard deviation of distances from the unity line in Fig. 6 is 0.6 ppm and maximum distance 1.9 ppm. Differences of this magnitude may introduce biases in the data end product, suggesting the need of a careful consideration of the data selection and classification procedure.

Summary and conclusions

The CO_2 annual cycles showed differences between the sectors. Air masses from the North and

Table 2. Degree of agreement between trajectory and wind direction based sector classifications for all trajectories.

Confidence level of sector classification according to trajectory	Percentage of successful sector classifications according to wind direction (all cases)	Percentage of unsuccessful sector classifications according to wind direction (all cases)
90%	567/2779 × 100 = 20%	215/2779 × 100 = 7.7%
75%	813/2779 × 100 = 29%	549/2779 × 100 = 20%
50%	1330/2779 × 100 = 48%	1226/2779 × 100 = 44%

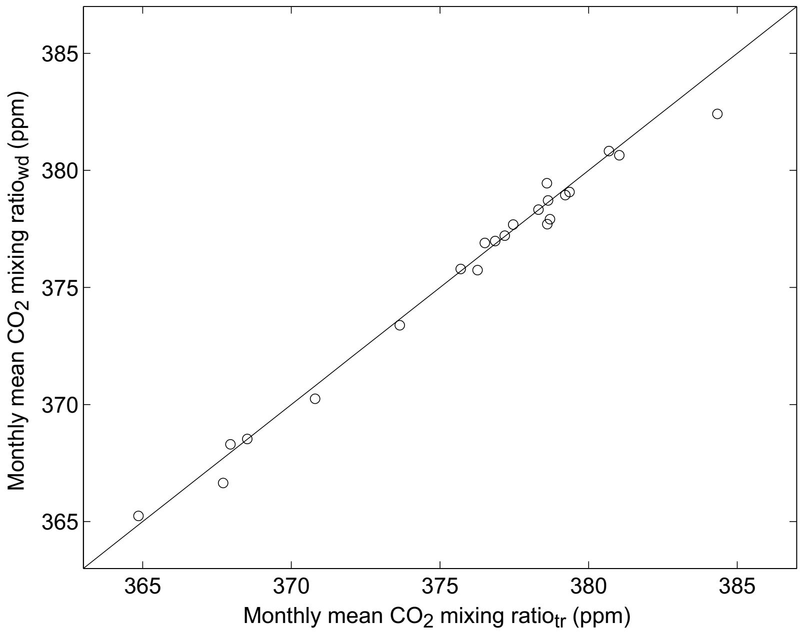

Fig. 6. Monthly mean CO_2 mixing ratios at Pallas according to different source sectors. 'wd' refers to wind direction based source sector determination and 'tr' to trajectory based source sector determination. Line is one-to-one line, which the points should meet if 'wd' and 'tr' based sector determinations agree for each 3h value comprising one month.

West sectors showed smaller amplitudes, affected possibly by long-range-transported marine air. Higher amplitudes were observed in air masses from the more continental South and East sectors. The CO_2 mixing ratio in air masses from the South sector was higher during late autumn than those from other sectors. In fact, the annual cycles corresponding to the North and West sectors were so similar that the division into two sectors can be impugned. On the other hand, the characteristics of the regions are quite different. The arctic source region is isolated and climatic conditions different from the Atlantic marine source region. For the needs of users of the data it probably is better to classify them as different sectors.

Classifying the source sector according to wind direction or trajectories resulted in significantly different monthly mean mixing ratios for CO_2. Trajectories are basically a more relevant method for a source area definition, since paths of air parcels can be followed several days backward and source area definition can be based on this information, not just the last hour before observation. Source areas do not need to be sectors but rather regions with similar characteristics. However, there are some problems in using trajectories together with long-term mixing ratio monitoring. For example, should the information about the boundary layer height be included in the analysis and how, and how should the gaps be filled since usually only few trajectories are provided per day while mixing ratio values are often averaged on an hourly basis. Furthermore, the source sector definition is not straightforward and may lead to a very low number of acceptable observations. If the required limit is 90% of the trajectory length inside the same sector, only 20% of the trajectories are accepted according to this work. A wind direction can, to some extent, be used instead of trajectories (*see* also Haagenson and Sperry 1989). However, only less than half of the set of 2779 trajectories can be classified and furthermore, classified correctly using the wind direction.

Trajectories seem to be a more suitable way of classifying carbon dioxide observations at Pallas. However, by using a wind direction-based sector classification we were able to identify different characteristics between the source sectors. Therefore, as long as continuous trajectory records are not available, it is reasonable to classify CO_2 observations according to wind direction by using four distinct source sectors.

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