Adaptive selection of Diurnal Minimum Variation: a statistical strategy to obtain representative atmospheric CO_2 data and its application to European elevated mountain stations

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Abstract. Critical data selection is essential for determining representative baseline levels of atmospheric trace gases even at remote measurement sites. Different data selection techniques have been used around the world, which could potentially lead to reduced compatibility when comparing data from different stations. This paper presents a novel statistical data selection method named Adaptive Diurnal Minimum Variation (ADMV) based on CO_2 diurnal patterns occurring typically at high elevated mountain stations. Its capability and applicability were studied on records of atmospheric CO_2 observations at six Global Atmosphere Watch stations in Europe, namely, Zugspitze-Schneefernerhaus (Germany), Sonnblick (Austria), Jungfraujoch (Switzerland), Izaña (Spain), Schauinsland (Germany), and Hohenpeissenberg (Germany). Three other frequently applied statistical data selection methods were included for comparison. Among the studied methods, our ADMV method resulted in lower selection percentages with lower maxima during winter and higher minima during summer in the selected data, which can be understood as a better representation of the lower free troposphere. The measured time series were analyzed for long-term trends and seasonality by seasonal-trend decomposition technique. Compared with unselected data, mean annual growth rates of all selected datasets were not significantly different, except for the data recorded for Schauinsland. However, clear differences were found in the annual amplitudes as well as the seasonal time structure. Based on correlation analysis, results obtained by ADMV selection showed a better representation of the lower free tropospheric conditions.

1 Introduction

Continuous in situ measurements of greenhouse gases (GHGs) at remote locations have been established since 1958 (Keeling, 1960). Knowledge of background atmospheric GHG concentrations is key to understanding the global carbon cycle and its effect on climate as well as the GHG responses to a changing climate. A crucial issue when using data from remote stations remains the identification of time periods that are representative of larger spatial areas and their differentiation from periods influenced by local and regional pollution. If these two regimes are well disaggregated, the available datasets can represent more reliable information about long-term changes of undisturbed atmospheric GHG levels or be used to investigate local and regional GHG sources and sinks when specifically analyzing the deviations from the baseline conditions. In this study, the baseline conditions refer to a selected subset of data from the validated dataset, representing well-mixed air masses with minimized short-term external influences (Elliott, 1989; Calvert, 1990; Balzani Lööv et al., 2008; Chambers et al., 2016).

Measurement results depend on sampling methods, analytical instrumentation, and data processing. Validated data (labeled as VAL in this study to differentiate from the selected data) are usually obtained after signal correction, for example, due to interferences from other GHGs such as water vapor, calibration accounting for sensitivity changes of the analyzer, and validation based on plausibility checks. Data selection starts with validated data and identifies in subsequent steps a final subset of the validated data set based on predefined criteria for specific qualities such as representativeness. With a particular focus on CO_2 in this study, it is commonly accepted that data selection methods can be categorized into meteorological, tracer, and statistical selection methods (Ruckstuhl et al., 2012; Fang et al., 2015).

Meteorological data selection makes use of the meteorological information at the measurement sites, which provides valuable information about the surrounding environment as well as air mass transport (Carnuth and Trickl, 2000; Carnuth et al., 2002). Forrer et al. (2000), Zellweger et al. (2003), and Kaiser et al. (2007) intensively studied the relationship between measured trace gases (such as O_3, CO, and NO_x) and meteorological processes at Zugspitze, Jungfraujoch, Sonnblick, and Hohenpeissenberg. For CO_2, the most common parameters applied in the literature are wind speed and wind direction. They can provide information on critical variations at stations with sources and sinks in their vicinity, while these parameters are less suited at stations in largely pristine environments. For example, Lowe et al. (1979) performed a pre-selection on the CO_2 record at Baring Head (New Zealand) during the southerly wind period only (clean marine air). In addition, Massen and Beck (2011) found that the CO_2 versus wind speed plot can be valuable for baseline CO_2 estimation without a local influence of continental measurements. Besides, fixed time window selection has been widely used, by selecting data in a certain time interval of the day based on local and mesoscale mechanisms of air mass transportation. For selecting well-mixed air at elevated mountain sites, night-time is usually chosen with a special focus on the exclusion of afternoon periods due to the influence of convective upward transport (Bacastow et al., 1985). Brooks et al. (2012) also limited their mountaintop CO_2 results in the Rocky Mountains (USA) by "time-of-day" from 0 a.m. till 4 a.m. local time (LT) to increase the likelihood of sampling the free tropospheric environment at the station. Apart from this, modeling techniques such as backward

trajectories are very helpful for analyzing in detail the origins and transport processes of air masses arriving at the station (Cui et al., 2011). Uglietti et al. (2011) focused on the origins of atmospheric CO_2 at Jungfraujoch (Switzerland) by the FLEXible PARTicle dispersion model (FLEXPART). Using tracers, data selection can be performed by investigating the correlations between the air components of interest. Many tracers have been applied and compared with CO_2. Threshold

5   limits of 300 ppb for CO and 2000 ppb for CH_4 were defined by Sirignano et al. (2010) to perform a regional analysis of CO_2 data at Lutjewad (the Netherlands) and Mace Head (Ireland). Similar approaches with black carbon and CH_4 were performed by Fang et al. (2015) at Lin'an (China). Moreover, Chambers et al. (2016) developed and applied a data selection technique to identify baseline air masses using atmospheric radon measurements at the stations Cape Grim (Australia), Mauna Loa (Hawaii, USA), and Jungfraujoch (Switzerland).

10   Unlike most of the methods mentioned above, which require additional data or advanced transport modeling, statistical data selection only relies on the time series of interest and typically investigates the variability of signal. It is usually assumed that the most representative CO_2 data are found during well-mixed conditions revealing small variations in time (Peterson et al., 1982) and in space (Sepúlveda et al., 2014). For continuous measurements, it is possible to investigate within-hour and hour-to-hour variability in the datasets. The within-hour variability is often expressed as the standard deviation of the measured

15   data within 1 h. The hour-to-hour variability compares the differences between hourly averaged concentrations either during a certain time period, or from one hour to the next. Pales and Keeling (1965) marked ambient data as "variable" when the within-hour variability for the air sample is significantly larger than the within-hour variability for the reference gas. Consequently, they only selected CO_2 data in "steady" conditions for 6 h or more. Besides, Peterson et al. (1982) also rejected sampled CO_2 data values for adjacent hours when the hour-to-hour variability exceeded 0.25 ppm. Thoning et al.

20   (1989) combined these two strategies using an iterative approach by selecting data according to deviations of daily averages from a spline curve fit. Ruckstuhl et al. (2012) developed a method based on robust local regression, called Robust Extraction of Baseline Signal, to estimate the baseline curves generalized for atmospheric compounds, which is available in the R package IDPmisc (Locher and Ruckstuhl, 2012).

The present study focuses on the comparison of results from statistical data selection methods and the Adaptive Diurnal

25   Minimum Variation (ADMV). The ADMV is seen as a possible alternative to already known data selection methods as discussed above. By applying ADMV to the atmospheric CO_2 records from six European elevated mountain stations, the selection results are compared with those derived from three other statistical data selection methods. To investigate the potential influences of the data selection method on trend and seasonality, further analyses focus on the decomposition of validated and selected data-sets in trend and seasonal components. Finally, differences between ADMV and other data

30   selection methods were assessed by correlation analysis.

2 Methods

2.1 CO_2 measurements at elevated European sites

CO_2 measurements from six selected European mountain stations (see Fig. 1) within the Global Atmosphere Watch (GAW) network were used to test the data selection algorithms. The data have been taken from mountain stations due to their remote location, which results in them being subjected to limited anthropogenic influence and having increased representativeness. Three high-alpine measurement sites were included: Zugspitze-Schneefernerhaus (*ZSF*, DE, 47°25′ N, 10°59′ E, 2670 m a.s.l.), Jungfraujoch (*JFJ*, CH, 46°33′ N, 7°59′ E, 3580 m a.s.l.), and Sonnblick (*SNB*, AT, 47°03′ N, 12°57′ E, 3106 m a.s.l.). They are often above the planetary boundary layer (PBL) and thus exposed to free and assumed clean lower tropospheric air masses, but are periodically influenced by regional emissions from lower altitudes. Additionally, to test data selection for a less remote environment, CO_2 measurements from Schauinsland (*SSL*, DE, 47°55′ N, 7°55′ E, 1205 m a.s.l.) at a clearly lower elevation in the mid-range Black Forest were investigated. Data selection was also applied to three recently started CO_2 time series from different sampling heights above ground at a tall tower at the Hohenpeissenberg observatory (*HPB*, DE, 47°63′ N, 11°01′ E, 934 m a.s.l.), located in the northern foothills of the Alps. Henne et al. (2010) presented a method of categorizing site representativeness based on the influence and variability of population and deposition by the surface fluxes. *JFJ* and *SNB* are classified as "mostly remote," while *ZSF* is considered as "weakly influenced, constant deposition," and *SSL* and *HPB* are considered as "rural" (Henne et al., 2010). Finally, Izaña station on Tenerife Island in the North Atlantic (*IZO*, ES, 28°19′ N, 16°30′ W, 2373 m a.s.l.) was chosen as a reference for comparison due to its location above the subtropical temperature inversion layer, which means that the station is rarely affected by any local or regional CO_2 sources and sinks (Gomez-Pelaez et al., 2013).

For this study, hourly data were used consistently, unless otherwise indicated. The validated CO_2 hourly averages from all stations were downloaded from the World Data Centre for Greenhouse Gases (http://ds.data.jma.go.jp/gmd/wdcgg/). Data with higher time resolution required for this study were provided directly by the station investigators. In addition, the time stamp was defined as the beginning of the averaging interval. Descriptions of the sampling elevation and time period of available data are given in Table 1. Further information on each station can be found in the report by Schmidt et al. (2003) for *SSL*, Gilge et al. (2010) for *HPB* and *SNB*, Gomez-Pelaez et al. (2010) for *IZO*, Risius et al. (2015) for *ZSF*, and Schibig et al. (2015) for *JFJ*. Practical data selections and analyses in this study were performed in the R Statistical Environment (R Core Team, 2017).

2.2 ADMV

ADMV is a tool for automated and systematic analysis of diurnal CO_2 cycles at elevated mountain stations in order to select consecutive time sequences with minimum variation, which can be regarded as representing well-mixed air conditions. Even though such measurement sites are remotely located, the CO_2 levels are still influenced by local sources and sinks. For example, at ZSF, these can be characterized by anthropogenic CO_2 sources, detectable especially in winter during the day,

whereas in summer the convective upwind transport results in a strong impact of air masses with depleted CO_2 concentrations due to photosynthesis at lower altitudes. Plant respiration activities, which may contribute small amounts, are primarily not visible in the convective upwind air masses (which arrive at mountain sites predominantly in the afternoon). Although high elevated mountain stations do not have vegetation in their surroundings, mountain stations at lower altitudes

5   but still in the vegetation zone may be influenced by plant respiration, especially at night. This points out the importance of finding a certain diurnal time window representing the most stable and representative CO_2 level, which in turn can be used for selecting representative data. However, the duration of this time window during the day varies with the season and from day to day because of variations in the dynamics of transport to the site (e.g., Birmili et al., 2009; Herrmann et al., 2015). In summer, larger variabilities in the CO_2 signal are observed due to more prevalent convective boundary layer air mass

10   injections influencing the diurnal pattern, resulting in shorter periods of stable conditions, whereas in winter, significantly longer stable periods occur. In winter, no upwind air masses with depleted CO_2 levels due to photosynthesis by vegetation are recorded. To receive as much representative data as possible, it is desirable to select the time window dynamically. ADMV is constructed to select a subset from the measured data, being best representative for baseline conditions with an adaptive selected time window specific for every day.

15   The algorithm is based on two basic assumptions. First, air masses measured at altitude stations contain well-mixed air, closest to baseline levels, within a certain time window of several hours during the day. For the elevated mountain stations discussed in this paper, this time interval is around midnight. Different diurnal patterns are apparent at each station, so the selected time window should be adjusted accordingly. Second, it is assumed that the real baseline conditions are not subject to local influences and thus represent air masses originating only from the uninfluenced lower free troposphere. This

20   indicates that the variability of the measured CO_2 signal should be minimal within this selected time window. The methodological steps of ADMV are introduced in detail below in the two sections *starting selection* and *adaptive selection*.

2.2.1 *Starting selection*

For a given validated hourly dataset, ADMV starts data selection by finding a *start time window* for all days. The standardized selection procedure for the *start time window* results from site-specific parameters. This time interval is set as

25   the most stable period from the diurnal variation. The step is referred to as *starting selection*. It begins by analyzing the mean diurnal cycle of the data input.

Step 1: Detrending is done by subtracting a 3-day average for each day, including the neighboring two days. It is the shortest possible time window to remove sudden changes in the time series related to the previous and posterior days while preserving the diurnal pattern.

30   **Step 2**: The overall mean diurnal variation, $\bar{d}_i$ ($i = 0 \ to \ 23 \ h$), is calculated from the complete set of detrended data.

Step 3: The standard deviations s_{Δ_j} from the overall mean diurnal variation $\bar{d}_i$ are calculated on a moving window Δ_j ($j = 6 \ h$). To be able to place a full set of 24 moving time windows over the overall mean diurnal variation, time windows

across midnight (e.g., 6 h from 11 p.m. to 4 a.m. LT) are also included, that is, its first j hours are appended to the end of the 24 h in the overall mean diurnal variation. The time window with the smallest standard deviation is selected as the *start time window*.

Result: The *start time window* $[i_{start}, ..., i_{end}]$.

5   With the focus on elevated mountain stations, *starting selection* is purposely designed with the moving window Δ_j of 6 h, and the starting hour i_{start} to be between 6 p.m. and 5 a.m. LT for this study. For other stations with possibly different diurnal patterns, *starting selection* can be adjusted accordingly. For instance, at urban stations or stations completely within the continental PBL, the *start time window* can be chosen based on the best mixing conditions, which often occur in the afternoon with a shorter moving window, when the PBL reaches its maximum depth after "ingesting" free tropospheric air

10   during its growth. Being aware that calculating the *start time window* from overall data could differ from the *start time windows* calculated by season, the overall generated *start time windows* have been compared with seasonal generated *start time windows* for highly elevated mountain stations (see Supplement S1.1). Because these differences are mostly minimal to moderate and this work aims at a methodical comparison under identical conditions, constant generation of *start time window* from overall data has been chosen.

15   **2.2.2 *Adaptive selection***

The second part *adaptive selection* is designed to determine the most suitable time window for each day, based on the data variability. The length of the *start time window* is adapted (expanding only) in both directions in time. *Adaptive selection* is performed on a daily basis, starting with the first day of the given data-set. The following steps only describe the *forward adaptive selection*. ADMV runs the *backward adaptive selection* in an analogous manner but going back in time.

20   **Step 1**: The mean molar fraction $\bar{x}_i$, standard deviation s_i, and the proportion of missing values $\pi_{missing}$ are calculated from data in the *start time window* $[i_{start}, ..., i_{end}]$.

Step 2: If $s_i \leq 0.3$ ppm (CO_2) and $\pi_{missing} \leq 0.5$, ADMV continues to advance in time to examine whether the next data point x_f can be included in the selected time window W with $f = i_{end} + 1$. Otherwise, it is considered that the *start time window* does not fulfill the assumptions. No data are selected for this day. One should proceed on to **Next Day**.

25   **Step 3**: The absolute difference between x_f and $\bar{x}_i$ is calculated, and the following threshold criterion is applied: $\left| x_f - \bar{x}_i \right| \leq \kappa \cdot s_i$, where κ is the threshold parameter. If this criterion holds, x_f is included in W and ADMV continues. Otherwise, ADMV is finished only with the *start time window* for this day and one should proceed on to **Next Day**.

Step 4: Mean $\bar{x}_W$ and standard deviation s_W for the new selected time window W are calculated. If $s_W \leq 0.3$ ppm (CO_2), ADMV continues with the next data point x_f while $f = f + 1$. Otherwise, ADMV is finished with the previous selected

30   time window and one should proceed on to **Next Day**.

Step 5: The new absolute difference between x_f and $\bar{x}_W$ is calculated, as well as the new threshold criteria. If condition $|x_f - \bar{x}_W| \leq \kappa \cdot s_W$ holds, x_f is included in W and ADMV goes back to **Step 4**. Otherwise, ADMV is finished for this day and one should proceed on to **Next Day**.

When selection for all days is finished, ADMV continues with *backward adaptive selection*. Afterwards, one should proceed on to **Result**.

Result: This is the final selected time window, which is a combination of $W_{forward}$ and $W_{backward}$ for the referring day.

The following limitations of the forward and backward expansions of the time window should be considered. ADMV always runs for no longer than 24 h including the *start time window*, namely, $f \leq 24 \cdot tr$, where tr is the time resolution in data points per hour of the input data. This sometimes results in an overlap of "selected" and "unselected" data for two consecutive days. We always label the data as "selected" once it has been selected by ADMV. The threshold parameter κ is the controlling factor of ADMV for the length of the selected time window. As κ increases, the length of the selected time window becomes larger. The value of 2 was chosen heuristically for this study as a compromise between selecting as many data points as possible and achieving the least data variability. Similar values of sensitivity-controlling parameters in other data selection methods can be found (Thoning et al., 1989; Sirignano et al., 2010; Uglietti et al., 2011; Satar et al., 2016). In **Step 2**, values of 0.3 ppm and 0.5 indicate the threshold values for s_i and $\pi_{missing}$. We denote them as $s_{i,threshold}$ and $\pi_{missing,threshold}$. It has been shown that less remote stations at lower altitudes require a larger value than 0.3 ppm because of different mixing conditions. When performing ADMV data selection at lower sites such as *HPB* and *SSL*, we recommend a higher $s_{i,threshold}$, such as 1.0 ppm instead of 0.3 ppm. However, throughout this study, we used the described parameter setting (0.3 ppm) for a methodical inter-comparison of selection methods at all stations. Potential influences of these parameter sizes ($s_{i,threshold}$ and tr) are discussed in Supplements S1.2 and S1.3.

2.3 Other statistical data selection methods for comparison

We compared ADMV with three statistical data selection methods. The first method named SI is based on "steady intervals" (Lowe et al., 1979; Stephens et al., 2013). Steady intervals, which are considered as baseline conditions, are defined by a standard deviation being lower than or equal to 0.3 ppm for 6 or more consecutive hours.

Second, we adopted a method applied by NOAA ESRL, which originated from Thoning et al. (1989). This selection routine has been applied specifically for measurements of background CO_2 levels at Mauna Loa. This method (referred to as THO) was applied as described on the website http://www.esrl.noaa.gov/gmd/ccgg/about/co2_measurements.html. The first step of THO examines the within-hour variability by selecting hours with hourly standard deviation less than 0.3 ppm. Second, it computes hourly averages and checks the hour-to-hour variability by retaining any two consecutive hourly values where the hour-to-hour difference is less than 0.25 ppm. The last step is based on the diurnal pattern (similar to ADMV), by excluding data from 11 a.m. to 7 p.m. LT due to transported air influenced by photosynthesis.

The last method compared is a moving average technique (MA). A moving time window of 30 days and a threshold criterion of two standard deviations from the moving averages were applied to discard outliers. Afterwards, new moving averages and new threshold criteria were calculated for data exclusion. This step is repeated until no more outliers can be found. A more detailed description can be found in Uglietti et al. (2011) and Satar et al. (2016).

2.4 Seasonal-trend decomposition STL

To analyze and compare the selected results from different data selection methods as well as the original validated data-sets, we applied the seasonal-trend decomposition technique based on locally weighted regression smoothing (Loess), named STL (Cleveland, 1979; Cleveland et al., 1990). STL has been widely used on measurements of atmospheric CO_2 and other trace gases (Cleveland et al., 1983; Carslaw, 2005; Brailsford et al., 2012; Hernández-Paniagua et al., 2015; Pickers and Manning, 2015). It decomposes a time series of interest into a trend component T, a seasonal component S, and a remainder component R, which allows separate detailed analyses and comparisons of trend and seasonality. Two recursive procedures are included in the STL technique: an inner loop where seasonal and trend smoothing based on Loess are performed and updated in each pass, and an outer loop that computes the robustness weights to reduce the influences of extreme values for the next run of the inner loop (Cleveland et al., 1990).

For this study, we used the implemented function `stl` in R (R Core Team, 2017). Owing to limitations of function `stl`, full time coverage of monthly data is needed in order to reduce the risk of large time gaps or unequal spacing (Pickers and Manning, 2015). All data results were first aggregated to monthly averages. Then, missing data were substituted by linear interpolation, using R function `na.approx` (Zeileis and Grothendieck, 2005). For the application of STL, two parameters need to be specified, which are the seasonal smoothing parameter $n_{(s)}$ ($s.window$ in function `stl`) and the trend smoothing parameter $n_{(t)}$ ($t.window$ in function `stl`). As $n_{(s)}$ and $n_{(t)}$ increase, the seasonal and trend components get smoother (Cleveland et al., 1990). For optimal compatibility in this study, the same parameters were chosen for all stations as $n_{(s)} = 7$ and $n_{(t)} = 23$, based on the recommendation of Cleveland et al. (1990). Another parameter combination of $n_{(s)} = 5$ and $n_{(t)} = 25$ was also tested according to Pickers and Manning (2015), but with no significant differences in results.

3 Results and discussion

3.1 *Start time window*

ADMV was applied to the validated hourly averages from all six stations with the parameter settings as described above. The detrended mean diurnal cycles were obtained with the *start time window* for each station by *starting selection* (see Fig. 2, for conventional mean diurnal plots see Supplement S2). The observed differences in the *start time windows*, as well as in the widths of the confidence intervals (gray shades in Fig. 2), can be explained by different site environments and thus differing data variabilities. The first subplot column (*HPB50*, *HPB93*, and *HPB131*), representing the three sampling heights at *HPB*,

shows similar detrended diurnal patterns with similar *start time windows*. The decreasing amplitude with increasing sampling height indicates that the higher the sampling inlet is above the ground, the less it is affected by the local surface fluxes. The three *start time windows* suggest that the most stable period at *HPB* occurs during the last few hours of a day, also including midnight. However, in contrast to all other stations covering at least a full year, *HPB* data are only from
5   September of 2015 to June of 2016. The results may not be fully comparable.

Regarding the second subplot column (*SSL*, *SNB*, and *IZO*), the *start time windows* can be found from midnight on or later in the morning. The *start time window* for *SSL* encompasses its diurnal maximum, indicating that data variability is considerably smaller in the early morning than in the afternoon because of its vicinity to the Black Forest region, which has strong influence due to local photosynthetic activities (Schmidt et al., 2003). A similar diurnal pattern can be found at *SNB*.
10   The influence of CO_2 sources is not as prominent as the effect of distant CO_2 sinks, since it is situated at the single summit peak of Hoher Sonnblick only surrounded by mountains and glaciers, with a negligibly small number of tourists, thus anthropogenic activities are minimal. *IZO* is a special case, since it is located on a remote mountain plateau on the Island of Tenerife above the strong subtropical temperature inversion layer. Even though the *start time window* is limited to 6 h, *IZO* presents an ideal mean diurnal cycle for data selection from a potentially much longer time window.
15   In the right subplot column, both *ZSF* and *JFJ* find their *start time windows* around midnight (with more hours after midnight). *ZSF* shows higher diurnal CO_2 amplitude than *JFJ*, but the two sites show similar diurnal patterns. For the choice of the *start time window* from the mean diurnal variation, relatively close or even local anthropogenic sources may influence the CO_2 at these two stations, possibly due to touristic influences.

3.2 Selection percentage

20   With the determined *start time windows*, ADMV selected the data for all stations (see Fig. 3). In addition, we calculated the percentages of ADMV selected data values among all values of the complete datasets for all stations, which are listed in the first column of Table 2. The higher the selection percentage is the more well-mixed air is measured at the station, which is assumed to be a representation of lower free tropospheric conditions. This holds especially for *IZO*. Because of this the greatest amount of accepted data points with 36.2% was found at this station. The sites with intermediate percentages are
25   *JFJ* (22.1%), *SNB* (19.3%), and *ZSF* (14.8%). For the three sampling heights at *HPB*, only 3.2% (50 m), 4.8% (93 m), and 6.2% (131 m) of the data were selected by ADMV. Finally, a similar percentage was found for *SSL* (4.0%), probably due to its higher data variability.

Since the stations were listed according to their altitudes, it was clear that all four selection percentages increase with altitude, indicating which indicated that measurements at higher altitudes could capture progressively well-mixed and hence
30   representative air. Therefore, linear least-squares regression was applied between the absolute altitude and the selection percentage for continental stations. *IZO* was on a remote island and therefore not comparable. This approach revealed a significant positive linear trend (see coefficient in Table 2). The related figure of linear regression can be found in Supplement S3.2.

To examine the characteristic growth of ADMV selection percentages during the selection process, we additionally calculated selection percentages after completing both the *starting selection* and *adaptive selection* steps mentioned in Section 2.2 (see Supplement S3.1). All of the results of percentages show an order of stations similar to that above, and the selection percentages increase steadily step by step for all stations.

5 The selection percentages of ADMV were compared again with those of the already mentioned statistical data selection methods SI, THO, and MA in Table 2, with the corresponding figure shown in Supplement S3.3. Since the selection percentages indicate not only the amount of data declared as representative but also show the characteristics of the selection methods, this criterion was used for further assessment. All other methods except for MA resulted in higher selection percentages for more highly elevated stations (*IZO*, *ZSF*, *SNB*, and *JFJ*) than for less elevated ones (*HPB* and *SSL*). ADMV

10 always performs the strictest in all cases. Based on the stepwise study of the selection percentages (see Supplement S3.1), such low percentages are due to the precise definition of the *start time window*. With *adaptive selection*, the selection percentages have grown but are still lower than those of the other methods. SI and THO, on the other hand, show differences between stations at high and low elevations. Compared with SI, THO is higher at stations at lower elevations, but lower at high ones. A major limitation of SI seems to be the requirement for consecutive hours, in our case of 6 h with 0.3 ppm

15 standard deviation threshold, which might be too restrictive for stations at lower elevations. However, this criterion results in a fairly large percentage for stations at high elevations. At *ZSF*, *SNB*, and *JFJ*, it results in the second largest, and even the largest in the case of *IZO*.

The highest selection percentages of approximately 80% were obtained with MA. However, owing to the minimal data variability of CO_2 measurements at *IZO*, the selection interval in MA becomes so small that the selection percentage

20 becomes considerably smaller than at all other stations. However, *IZO* obtains the largest selection percentages from all other selection methods. Thus, we conclude that MA does not work properly in the case of very well-mixed air (*IZO*). At all other stations, it is possible that MA declares too much data as representative. Therefore, MA was excluded from further analyses.

3.3 STL decomposed results

25 STL was applied to the validated datasets before and after selection with SI, THO, and ADMV, except for *HPB* due to its limited length of time (less than one year). Depending on data availability, STL was performed on CO_2 data from 2012 to 2015 at *SNB*, while data inputs at *SSL, IZO*, *ZSF*, and *JFJ* cover the whole period from 2010 to 2015. Figure 4 gives an overview of the decomposition in each component by STL. The following sections discuss the resulting components obtained by STL, namely the trend component over the observation period, the seasonal component and finally the

30 remainder component.

3.3.1 Trend component

From the trend component, the mean annual growth rate is estimated by linear regression (see Table 3). Based on the 95% confidence interval for the slope, a positive trend i.e. increasing CO_2 concentrations are observed. Owing to the overlap of the confidence intervals, differences in the mean annual growth rates among VAL and selected datasets at the same station

5   are all in good agreement. This indicates that the trend component is not influenced by the statistical data selection method, which agrees well with the finding of Parrish et al. (2012) from a study of baseline ozone concentrations that there were no significant differences of the long-term changes between the baseline and unfiltered datasets. Moreover, the following fact is observed for all sites except for *SSL*. Compared to unselected data (VAL), the mean annual growth rates based on selected data–sets are systematically higher approaching the growth rates at *IZO*. *IZO* can be considered as better representing the

10   lower free tropospheric conditions and agrees well with the mean annual absolute increase during last 10 years (2.21 ppm yr^{-1}) reported by WMO (2017a). The exception at *SSL* is probably caused by stronger local influences as a result of its lower elevation. Besides, the confidence intervals of the mean annual growth rates are always smaller after data selection, which improves the precision of trends.

3.3.2 Seasonal component

15   The resulting seasonal components show systematic differences between VAL and selected datasets. The mean monthly variations were calculated on a monthly scale over the entire period from the analyzed data. Figure 5 (a) and (b) present the results at stations *ZSF* and *IZO*. At most stations (except for *IZO*), the seasonal amplitudes have been substantially reduced compared to VAL (see also Fig. 4). At *ZSF*, the averaged peak-to-peak seasonal amplitude, defined as mean seasonal maximum minus seasonal minimum, drops the most by 18.9% from VAL with the ADMV selected dataset. An explanation

20   of this reduction is CO_2 signal exclusion from local sources and sinks by data selection. When taking a closer look at the monthly averages, lower CO_2 values are found in the selected datasets in the winter months from October to April, indicating that the CO_2 level is overestimated by VAL because of more dominant anthropogenic activities and no active vegetation. Higher values in the summer months from May to September explain underestimation of VAL due to intensified upward transport of photosynthetic signatures resulting from vegetation. Similar patterns can be found at stations *SSL*, *SNB*, and *JFJ*

25   (see Supplement S4). *IZO*, as expected by its location, always shows the smallest seasonal amplitude and nearly uninfluenced monthly results between VAL and selected datasets. Based on this consideration, it is very likely that the lower free troposphere will react with a delay to CO_2 concentration changes of effective sources and sinks on the ground, acting like an atmospheric memory.

A time delay of one month in the mean seasonal maximum is shown in Fig. 5 (a) at *ZSF* with selected datasets by SI and

30   ADMV (March), compared with the maximum from the validated data (February). In addition, a similar time shift can also be found by other selection methods at stations *SSL* (one-month delay from February to March by SI and ADMV) and *JFJ* (two--months delay from February to April by SI, THO, and ADMV). As for station *IZO* (April) in Fig. 5 (b) and station

SNB (March), the seasonal maxima stay the same. The magnitude of these delays may be related to mixing features in the lower free troposphere. Rapid changes are usually observed close to sources and sinks, e.g., from anthropogenic and biogenic activities. Thus, the higher the station is above the boundary layer, the later the maxima during the winter can be observed, because of the late response due to inhibited mixing conditions. However, this delay does not occur for the minima during the summer because of the very effective upward transport and more favorable mixing conditions at that time of year. Consequently, no changes in the seasonal minima are observed at all measurement sites, which is taken as an indicator of enhanced thickness of the mixing layer as good mixing conditions. Taking *ZSF* as an example, Birmili et al. (2009) observed low concentrations of particle number in winter and found it representative for the free tropospheric air by analyzing the annual and diurnal cycles. From spring on, the warmer it gets the higher the PBL goes. The intense vertical atmospheric exchange during summer months results in a daily air mass transport from the boundary layer to reach *ZSF* due to thermal convection (Reiter et al., 1986; Birmili et al., 2009). Thus there are optimal transportation and mixing conditions. Therefore after data selection, the timing of seasonal peaks better corresponds among the stations.

3.3.3 Remainder component

The remainder component contains data with external and random influences. It has characteristics of random noise, being basically different from site to site and statistically uncorrelated with the general signal of CO_2 concentrations in the lower free troposphere (Thoning et al., 1989). The standard deviation of the remainder component is taken here as a measure for external influences (see Fig. 4). Table 4 shows the calculated standard deviations from the remainder components at each station. Comparable results are derived from all selected datasets. *SSL*, as the lowest elevated station, exhibits the most variation. *IZO* with the smallest standard deviations in the remainder components proves to be the station least influenced by its surrounding environment. The three alpine measuring stations (*ZSF*, *SNB* and *JFJ*) exhibit intermediate variability. From this perspective, STL performs well to show the site characteristics. Consequently, the noise of the remainder components, given in Table 4, decreases with increasing altitude of the continental mountain stations, which is in inverse relation to the selection percentage (Table 2). *IZO* was excluded in both regressions against altitude because of its maritime character.

3.4 Correlation analysis

As mentioned above, data selection is defined here as an approach of extracting a group of data to be the best representative for the lower free troposphere. Consequently, the selected CO_2 data–sets from all stations should theoretically agree better among themselves. For validation of this, we took the combination of the trend and seasonal components from STL and examined the correlations between each pair of stations in a Pearson correlation matrix (see the upper panel of Fig. 6). The trend and seasonal components of all VAL and selected datasets were first compiled, and then Pearson's correlation coefficients were calculated assuming normal distribution of data examined by the Anderson Darling test (P < 0.05). The correlation matrices are shown for each type of dataset individually. Data used for correlation were chosen only when available at all stations (2012–2015). In general, most pairs show higher correlation coefficients with selected data from the

different selection methods, especially between the three Alpine stations (*ZSF*, *SNB*, and *JFJ*). This evaluation hence shows a similar result to the method presented by Sepúlveda et al. (2014) for identifying baseline conditions based on the correlation between distant measuring stations. Pairs including *IZO* after data selection by ADMV show a notable increase in the correlation coefficients, meaning a better coherence between the reference station *IZO* and the others. On the other hand, when selecting representative data more effectively, the results should contain less local and regional influences. Therefore, we compared the remainder components derived from STL pairwise to check whether the Pearson correlation coefficients decreased after data selection, as shown in the lower panel of Fig. 6. The number of insignificant correlations between the station pairings is the greatest for ADMV. For the only two coefficients significant at the 0.05 confidence level (*ZSF-SNB* and *ZSF-JFJ*), they drops largely from 0.75 to 0.48, and from 0.75 to 0.40, respectively, which cannot be observed by the other selection methods. This means that by ADMV the combination of trend and seasonal components correlate best and the remaining unselected data have the lowest correlation among the methods. If these two criteria are used to separate the representative part of the data from the unrepresentative part, the ADMV method produces the best results.

4. Conclusions and outlook

We presented a novel statistical data selection method, the ADMV, for CO_2 measurements at elevated GAW mountain stations. For validation and assessment of the data selection procedure, we applied the method to six CO_2 datasets measured at GAW mountain stations in the European Alps. The ADMV method resulted in an increasing percentage of data with growing altitude which is reasonable due to the underlying atmospheric dynamics. Comparing ADMV with three other well-known statistical data selection methods, all methods yielded rather consistent characteristics across different stations. Nevertheless, among all the methods, ADMV is the most restrictive in terms of the number of selected data in the overall datasets.

In addition, we applied the time series decomposition tool STL to all validated and selected datasets. All statistical data selection methods resulted in the same annual trend in terms of the 95% confidence interval from the validated data-sets, while the seasonal signal varied substantially with smaller seasonal amplitudes and delayed occurrences of seasonal maxima. We also presented an additional assessment of the proposed new method compared with the other statistical data selection methods based on correlation analysis. Both, higher correlation coefficients of the trend and seasonal components by STL and inversely lower coefficients of the remainder indicate a better performance of ADMV than the other methods SI and THO.

The presented method ADMV is useful for data selection of atmospheric CO_2 data representative of the lower free troposphere. It requires only data from a single measurement site. It is easily adjustable to the local conditions and it runs automatically. The method can also be applied to historical datasets. The results provide evidence that the proposed ADMV method confers the possibility of selecting data that are representative of CO_2 concentrations of a larger area of the lower free troposphere. This is an elementary prerequisite for application of the method to a larger number of different stations and

an essential step toward generalization. It directly supports the objective of GAW to extrapolate from a set of point measurements from single stations to a larger representative area or region in the lower free troposphere (WMO, 2017b). In future, there is a need to test whether such results could be used for additional tasks, such as ground calibration of satellite measurements. Finally, it would be extremely interesting to test whether this presented concept also holds in other regions and on other continents. Moreover, the issue of whether and how to include coastal stations in a systematic and practically generalizable approach for selecting representative data at GAW stations will be a particular concern.

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Table 1: Information of measured CO_2 data-sets at six GAW mountain stations.

Station (GAW ID)	Sampling elevation (a.s.l.)	Time period (yyyy.mm)	Data provider
Hohenpeissenberg (*HPB*)	984/1027/1065 m	2015.09–2016.06	DWD
Schauinsland (*SSL*)	1210 m	2010.01–2015.12	UBA-De
Izaña (*IZO*)	2403 m	2010.01–2015.12	AEMET
Zugspitze-Schneefernerhaus (*ZSF*)	2670 m	2010.01–2015.12	UBA-De
Sonnblick (*SNB*)	3111 m	2010.01–2015.12	UBA-At
Jungfraujoch (*JFJ*)	3580 m	2010.01–2015.12	Empa

Table 2: Selection percentages of selected data among all data by different data selection methods. The bottom shows the linear regression coefficients of stations' (*HPB* is represented by *HPB50*; *IZO* is excluded) altitudes and the selection percentages at a significance level of 0.05 (***).

Station ID	ADMV	SI	THO	MA
HPB50	3.2	13.9	21.7	79.8
HPB93	4.8	18.5	25.0	79.4
HPB131	6.2	21.3	27.3	79.8
SSL	4.0	17.9	25.4	83.2
IZO	36.2	82.2	56.0	60.5
ZSF	14.8	47.1	40.8	79.0
SNB	19.3	58.7	44.2	76.9
JFJ	22.1	62.1	46.3	77.6
Linear regression coefficient (γ^2)	0.996***	0.992***	0.985***	0.645

Table 3: Mean annual growth rates (ppm y $^{-1}$) with 95% confidence intervals from linear regression applied on the trend components by STL over 2010 to 2015, except for *SNB*. Data at *SNB* were decomposed over 2012 to 2015 due to missing data from 2010 to 2011 and thus shown in gray.

Station ID	VAL	SI	THO	ADMV
SSL	2.04 ± 0.09	1.89 ± 0.06	2.04 ± 0.06	2.03 ± 0.09
IZO	2.24 ± 0.03	2.26 ± 0.02	2.25 ± 0.02	2.25 ± 0.02
ZSF	2.13 ± 0.08	2.16 ± 0.05	2.17 ± 0.06	2.19 ± 0.06
SNB	2.02 ± 0.07	2.06 ± 0.06	2.06 ± 0.06	2.08 ± 0.04
JFJ	2.13 ± 0.03	2.15 ± 0.02	2.14 ± 0.02	2.14 ± 0.02

Table 4: Standard deviation of the remainder components by STL over 2010 to 2015, except for *SNB*. Data at *SNB* were decomposed over 2012 to 2015 due to missing data from 2010 to 2011 and thus shown in gray.

Station ID	VAL	SI	THO	ADMV
SSL	1.61	1.16	1.26	1.99
IZO	0.34	0.33	0.30	0.30
ZSF	0.89	0.75	0.72	0.73
SNB	0.66	0.56	0.55	0.70
JFJ	0.56	0.45	0.48	0.47
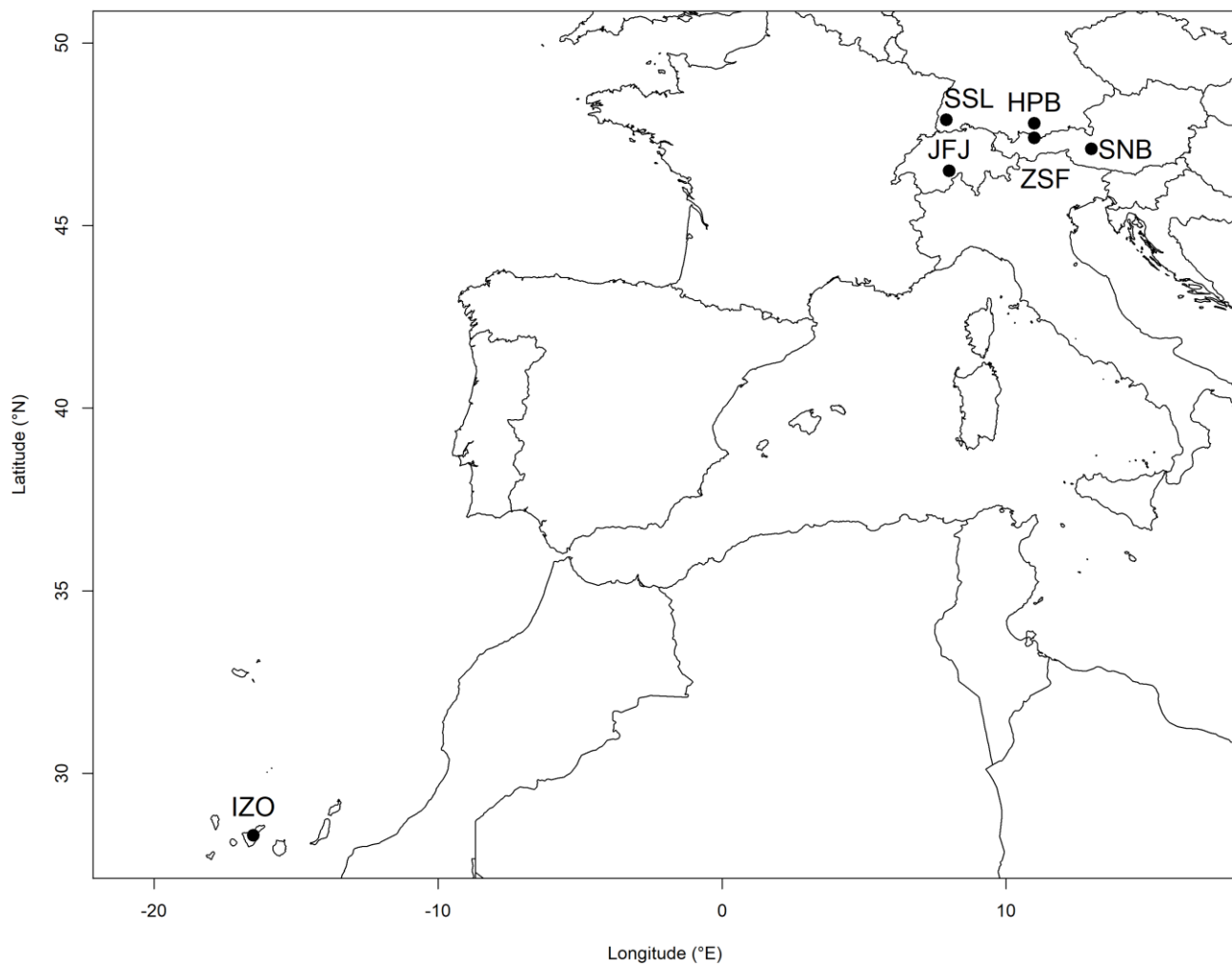
Figure 1: Locations of six European elevated mountain stations. Symbols from left to right stand for: IZO – Izaña, Spain; SSL – Schauinsland, Germany; JFJ – Jungfraujoch, Switzerland; HPB – Hohenpeissenberg, Germany; ZSF – Schneefernerhaus-Zugspitze, Germany; SNB – Sonnblick, Austria.
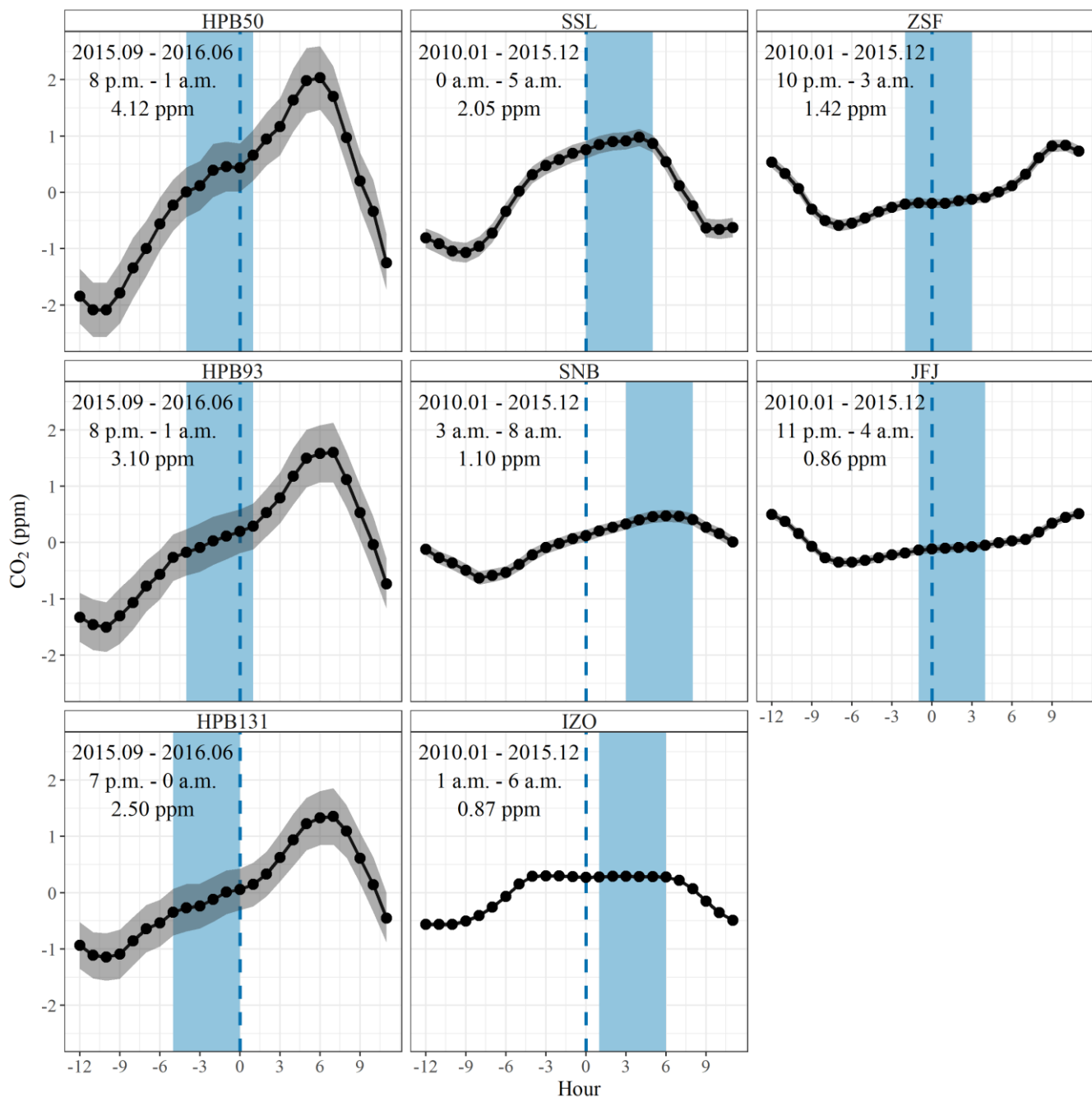
Figure 2: Detrended mean diurnal cycles of validated CO_2 datasets (black) with 95% confidence intervals (gray) from six GAW stations (hours in LT). Measurements at *HPB* are differentiated by the sampling heights (e.g., *HPB50* for 50 m a.g.l.). The covered time periods (top text), resulting *start time windows* (middle text, also in light blue shades), and mean diurnal amplitudes (bottom text) are shown in each subplot.
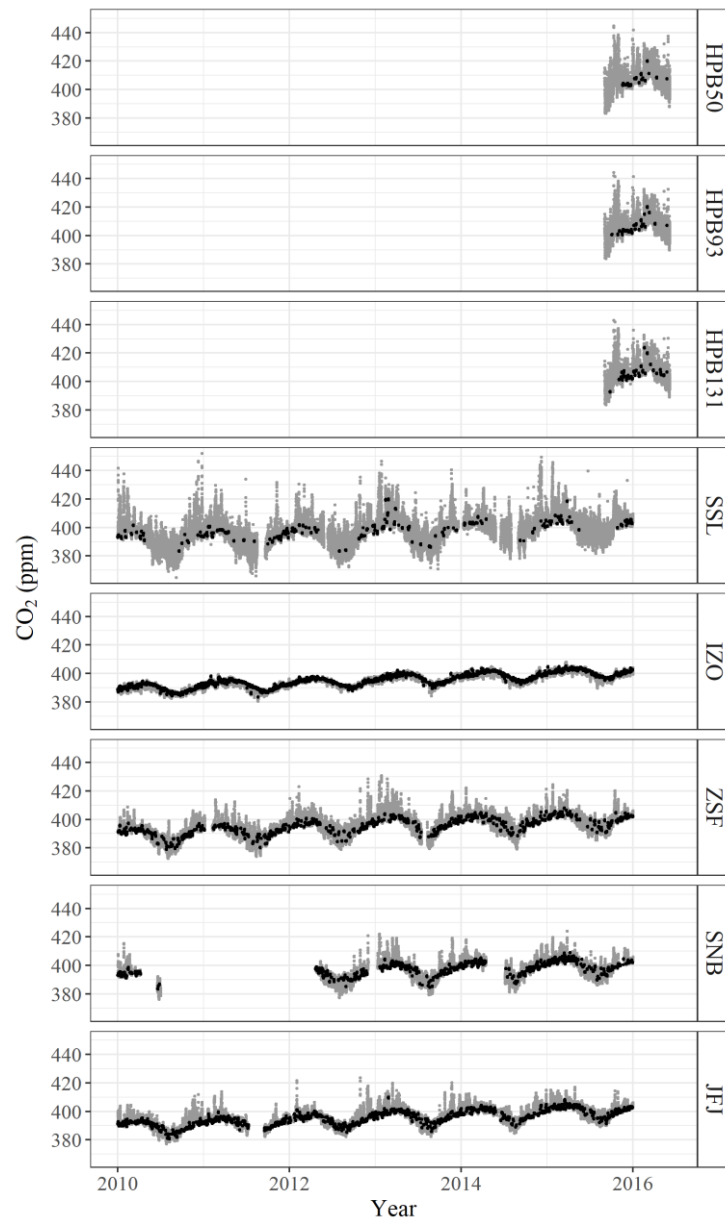
Figure 3: Time series plots of validated CO$_2$ datasets (gray), and selected data–sets by ADMV (black) at six GAW stations.
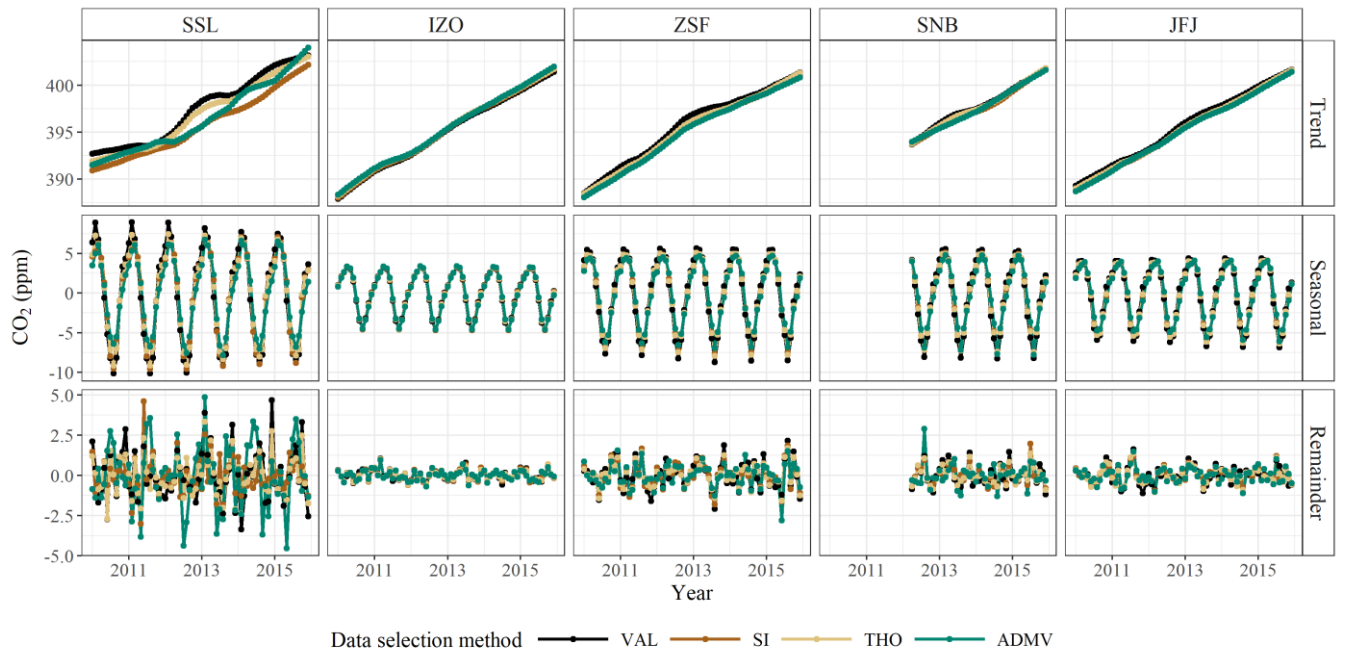
Figure 4: STL decomposition results from VAL (black), SI-selected (brown), THO-selected (yellow), and ADMV - selected (green) data-sets at five GAW stations.
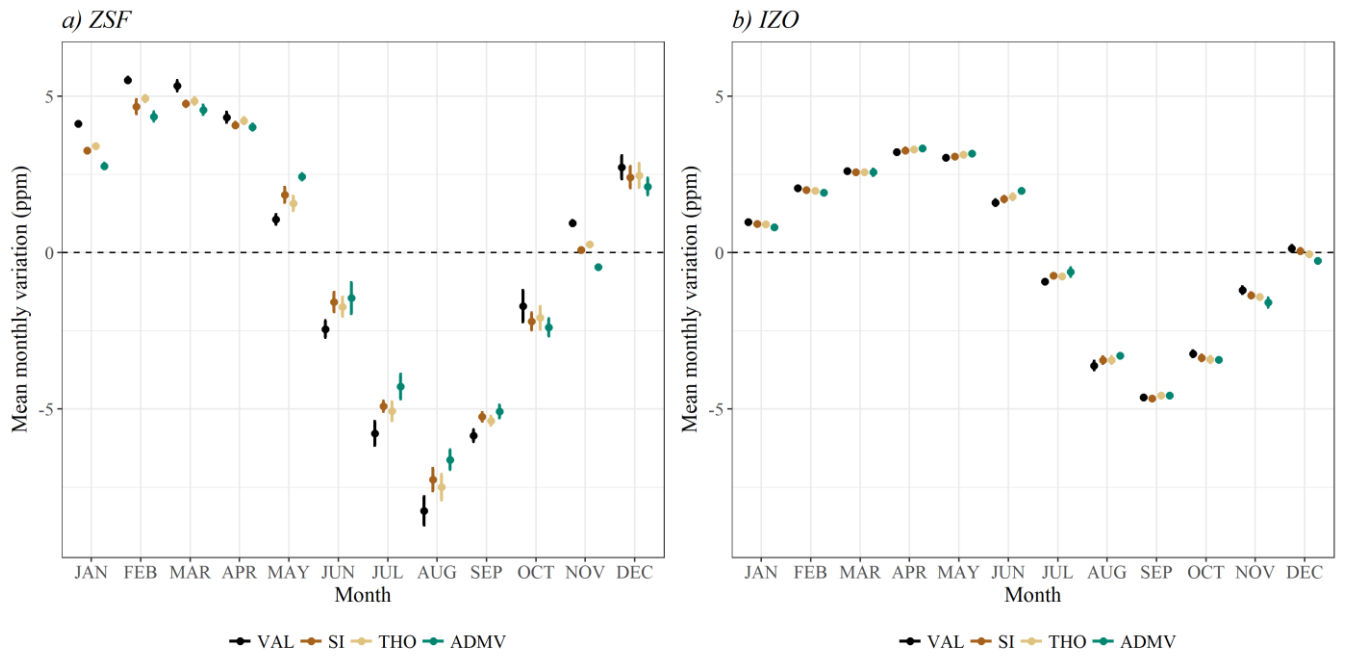
Figure 5: Mean monthly variation of the seasonal component decomposed by STL at a) *ZSF* and b) *IZO* over the whole period. For better visualization of the results of selection methods, dots have been separated horizontally equidistantly. The 95% confidence intervals are shown as error bars.
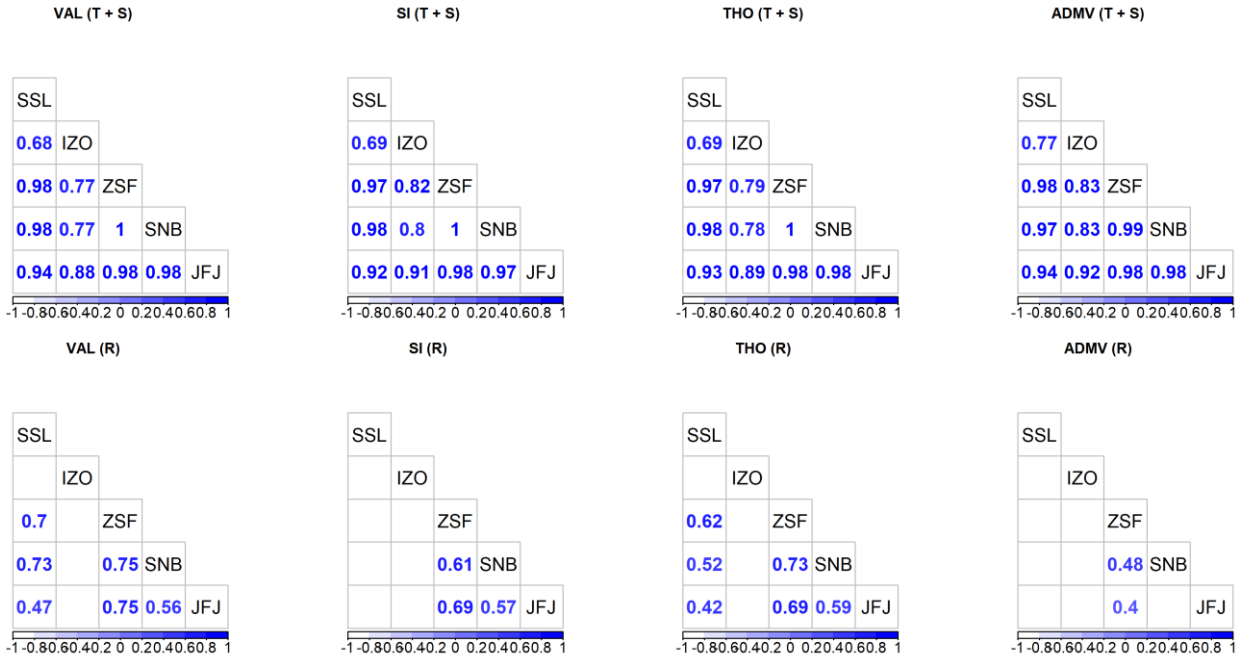
Figure 6: Pearson's correlation matrices of combinations of trend (T) and seasonal (S) components (upper panel), and only remainder (R) components (lower panel) at stations *SSL*, *IZO*, *ZSF*, *SNB*, and *JFJ* by different selection methods, as indicated on the top. The color scale reflects the strength of correlation. Correlations with no significant coefficient at the 0.05 confidence level were left blank.

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