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Investigating regional source and sink patterns of Alpine CO_2 and CH_4 concentrations based on a back trajectory receptor model

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Abstract

Background: The main purpose of this paper is to contribute to the improvement in the present knowledge concerning regional carbon dioxide (CO_2) and methane (CH_4) exchange as an essential step towards reducing the uncertainties along with bottom-up estimations of their global budget by identifying the characteristic spatial and temporal scales of the regional greenhouse gas fluxes. To this end, we propose a stepwise statistical top-down methodology for examining the relationship between synoptic-scale atmospheric transport patterns and mole fractions of the climate gases to finally receive a characterisation of the sampling sites with regard to the key processes driving the CO_2 or CH_4 concentration levels.

Results: The results of this study presented in this paper give detailed insights into the emission structures underlying the measurement time series by means of origin-related examinations of the Alpine CO_2 and CH_4 budgets. The time series of both climate gases from the atmospheric measurements carried out at the four high-alpine observatories Schneefernerhaus, Jungfraujoch, Sonnblick and Plateau Rosa form the basis for the characterisation of the regional CO_2 as well as CH_4 budget of the Alpine region as the focus area of the Central European study region. For the investigation area so outlined, the project identifies source and relative sink regions with influence on the Alpine climate gas measurements as well as their temporal variations. The therefore required combination of the measurements with the synoptic situation prevailing at the respective measuring time which carries the information about the origin of the analysed air masses is derived by means of a trajectory-based receptor model. The back trajectory receptor model is set up to decipher with high spatial resolution the most relevant source and sink areas, whereby the Alpine region is identified as a significant relative sink for CO_2 as well as for CH_4 concentrations all year long, whereas major European emitters show their impact during different seasons.

Conclusions: The reliable results achieved with this approach in connection with the encouraging model-internal uncertainty assessments and external plausibility checks lend credence to our model and its strength to illustrate dependably spatial–temporal variations of the relevant emitters and absorbers of different climate gases (CO_2 and CH_4) in high spatial resolution.

Keywords: CO_2 and CH_4 concentrations, High-alpine observatories, Back trajectory receptor model, Regional source and sink patterns

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Background

Carbon dioxide (CO_2) and methane (CH_4) represent the two most important greenhouse gases (except for water vapour) with a combined radiative forcing of 2.3 [±0.24] W/m^2 on the global average [18]. The unbroken increase in these two most notorious atmospheric greenhouse gases of over 120 ppm or 1.080 ppb above preindustrial levels has been unequivocally attributed to human emissions mainly coming from fossil fuel burning and land-use changes, while the oceans and terrestrial ecosystems somewhat attenuate this rise with seasonally varying strength [20, 33].

It is not only the climate gases time series themselves that matter regarding the unbroken increase in the atmospheric concentrations of both greenhouse gases concerning their preindustrial level (CO_2 by 40%, CH_4 by 150%) [18]. But more particularly, their interpretations with a special attention on the regional emission budget within the catchment area of measurement stations are of high scientific as well as sociopolitical interest. Only the knowledge about regional emission structures provides a sound understanding of the regional climate gases' budgets and therefore a valid detection of varying contributions in the catchment area of the measurement sites.

The derivation of such variables of a climate political dimension from high-precision measurement time series of climate gases on whose basis efficient emission reduction actions can be verified and adapted if necessary, is anything but trivial and requires the differentiated breakdown of the measurements by their origin. The interaction of mankind and biosphere as emitters or absorbers in connection with the long atmospheric lifetime—especially of CO_2—is decisive for the complexity of this task. The long lifetime of climate gases once emitted together with the interference of the anthropogenic emissions—here burning of fossil fuels as well as land-use changes and livestock farming have primarily to be mentioned—and the seasonal carbon cycle of the biosphere, just like natural biogeochemical cycles, prevent atmospheric measurement time series of the climate gases from immediately providing information about changes of the regional emission situation [20].

The periodic seasonality of the natural global cycles as well as the above-mentioned secular trends superimpose some short-term fluctuations on the CO_2 and CH_4 concentrations reflecting the influence of regional climate gas emitters and absorbers. In this context, the transient CO_2 and CH_4 components prompted the development of different methods specifically designed to identify with high spatial resolution the most relevant regions appearing as CO_2/CH_4 sources or relative sinks within the catchment area of a receptor site [1]. This kind of approaches set the short-term fluctuations in the climate gas concentration measured at a particular station in relation to the simultaneous synoptic conditions captured by statistical analyses of back trajectories [36]. For the reconstruction of dynamic processes in the atmosphere on the synoptic scale, trajectories from the dispersion and transport modelling have established themselves as reliable tool. Air mass trajectories give an approximation of the path that air parcels have covered over a period of time, thereby carrying a specific history due to the crossed-over regions with them [13]. The simulation of trajectories can therefore give detailed insights on how the emission situation affects the climate gas concentrations of the considered observatories. On the basis of meteorological fields out of numerical weather prediction models, trajectories track the movement of an air package in space and time, thereby indicating flow patterns. Trajectories calculated from a measurement site backward in time thus give information about the transport pathways and potential source regions of the detected air masses. Thereby backward trajectories infer geographic regions that contribute to pollution events and enable detailed insights into source–receptor relationships within the catchment area of the target station [30, 31]. As back trajectories cross locations along their route where the measured concentration anomalies were brought about, these combinations have been applied to study source–receptor relationships without converting atmospheric greenhouse gas concentrations (ppm/ppb) into emission data ($(kg\ m^2)/s$) but taking concentration anomalies as a proxy of sources and relative sinks [1].

One representative of the receptor-oriented methods is back trajectory receptor models calculating the movements of air parcels from the receptor site backward in time whereby the influence of crossed source and sink regions can be traced back by means of the resulting pathways. The basic assumption to which the trajectory-based receptor model is subject is that the air represented by backward trajectories is affected by the change of the climate gas concentration while passing a grid cell with relevant sources or sinks, such that the changes are effectively transported to the receptor [7]. The various forms of different types existing within the category of back trajectory receptor models, such as potential source contribution functions, gridded frequency distributions, concentration field analysis, residence time weighted concentration as well as concentration weighted trajectory fields (CWT), have in common that they indicate possible origins of measured air parcels on a grid, discretising the land surface and different height levels [9]. From that results the advantage of providing the spatial distribution of potential source and sink areas contributing to the measurements at the receptor site. Thus, with the back trajectory receptor models regional anthropogenic as well as biospheric point sources can be identified provided that the calculation has taken into account that the majority of the trajectory endpoints are all found surrounding the receptor location where all the back trajectories converge [32]. One formula that considers the increasing number of trajectory endpoints near the grid cell of the receptor site is the CWT approach [9]. This is done by normalising the source intensity of the grid cells by the trajectory residence time. Due to this amendment, the CWT approach is able to avoid the potential false identification of sources close to the receptor where the longest residence time could deceptively lead to the assumption of a greater influence on the concentrations measured at the receptor site. In addition, the trajectory residence time in the grid cells of the CWT is weighted by the concentration of the trace gas measured on arrival of each trajectory. The combined consideration of the detected concentration levels and the higher frequency of trajectory endpoints near the receptor in the model equation makes the CWT method a reliable and precise tool for source attribution studies [9].

The primary aim of our study is to give detailed insights into the emission structures underlying the measurement time series to examine carbon dioxide (CO_2) and methane (CH_4) budgets of the Alpine region by identifying the source and sink regions with influence on the Alpine climate gas measurements as well as their temporal variations. The required combination of the measurements with the synoptic situations prevailing at the respective measuring time intervals which gives the information about the origin of the recorded air masses is derived by means of the CWT approach. The highly precise climate gas time series of the Alpine observatories adjusted by long-term trend as well as the influence of seasonality from the biosphere on the one hand and the centroid tracks of the particle dispersion modelling arriving at the respective measuring times at the receptor locations on the other hand form the input data for the receptor model. Detailed emission inventories are not required for the application of receptor models.

Using this methodology, relevant sources and sinks with influence on the CO_2 and CH_4 concentrations of the years 2011–2015 as well as the temporal variability of the emitters and absorbers recorded at the Alpine observatories are sought. Beneath the identification of their seasonal and year-wise occurrence, an assessment to what extent the method applied here and the results obtained with it allow for reliable conclusions on emitters and relative sinks combined with their temporal variability. Thereto, the model's internal uncertainty is estimated and external plausibility checks are carried out by comparisons with results from the inverse modelling of climate gas fluxes and concentrations. In the end, the question has to be addressed whether the methodology of our study can be considered well capable of reliably detecting climate gas-specific source and sink regions with influence on the measurements at the Alpine receptor sites. Or in other words: How accurately and with what potential limitations can the model map spatiotemporal variations of the relevant emitters and absorbers of different climate gases (CO_2 and CH_4) measured at high-alpine observatories?

Study area

One promising method to adequately analyse the regional CO_2 and CH_4 budgets of an investigation region focuses on the time series of climate gases measured at high-situated observatories with a huge catchment area far away from local emission sources (potential impacts of the—even at these exposed sites—remaining local sources from tourism or machines as for example cableways were excluded carefully by the scientists responsible for the measurements). Due to their exposed position, these sites are particularly suited for investigations on the sources of climate gases, since they capture the state of the lower free troposphere just as the long-distance transport of air masses and aerial admixtures. These perfect circumstances are met in Central Europe in particular at the observatories in the Alpine high mountain region. Here, measurements are set up to adhere to high precision and quality assurance standards corresponding to their supra-regional significance. In particular, the requirements of the international program for atmospheric monitoring Global Atmosphere Watch (GAW) of the United Nations' World Meteorological Organisation (WMO) are met. The GAW monitoring program is responsible for the measurement of the physical and chemical state of the atmosphere within the UN/WMO Global Climate Observing System (GCOS). The central objective of GAW is to build up a global database of highly precise and hence compatible measurement data allowing a coherent worldwide analysis of atmospheric concentrations. For ensuring the required data quality objectives, which are less than ± 0.1 ppm CO_2 in the northern hemisphere and ± 2.0 ppb CH_4 for repeated inter-comparison measurements, GAW measurements are embedded in a scientific framework for quality assurance. The actual requirements for CO_2 and CH_4 measurements are entirely described in the GAW Report No. 242 [34].

The indicated data quality in connection with the high representativity of the measured climate gas concentrations certifies ideal conditions for the project objective to the measurements of the high-alpine observatories. Accordingly, the investigations of the CO_2 and CH_4 budgets introduced in this report are based on the measurement time series of the following four high-alpine measuring stations (see Table 1) which guarantee the widest possible cover of the Alpine region as core area of the Central European study area from the southwest up to the northeast (see Fig. 1):

- Environment Research Station Schneefernerhaus (UFS)
- Sphinx-Observatory Jungfraujoch (JFJ)
- Sonnblick Observatory (SOB)
- Observatory Plateau Rosa (PRO).

Decisive for the choice of exactly these four measuring stations (see Table 2 for detailed information about their measuring instruments) as database is—beside the guarantee of the highest data quality according to the GAW standards—in particular the supra-regional representativity of measurements conducted there. This is ensured

Table 1 Overview of the four high-alpine observatories whose climate gases' time series form the basis for the characterisation of the Alpine CO_2 and CH_4 budget

Name of the receptor site (abbreviation)	Country	Latitude	Longitude	Height (amsl)
Environment Research Station Schneefernerhaus (UFS)	Germany	47.42° N	10.98° E	2650 m
Sphinx-Observatory Jungfraujoch (JFJ)	Switzerland	46.55° N	7.98° E	3450 m
Sonnblick Observatory (SOB)	Austria	47.05° N	12.96° E	3106 m
Observatory Plateau Rosa (PRO)	Italy	45.93° N	7.70° E	3480 m

Fig. 1 Map showing the four high-alpine observatories (blue triangles) considered for the characterisation of the Alpine (red line) CO_2 and CH_4 budgets

Table 2 Overview of the instruments for measurement of CO_2 and CH_4 on the four high-alpine observatories

Site	Greenhouse gas	Model	Manufacturer	Measuring principle
Hoher Sonnblick	CO_2	Uras14 (2000–2012) G2301 (2012–…	ABB Picarro	NDIR
Jungfraujoch	CO_2	G1301 (2010–2011); G2401 (2011.09.16–…)	Picarro Picarro	CRDS
Plateau Rosa	CO_2	Ultramat	Siemens	NDIR
Zugspitze/Schneefernerhaus	CO_2	HP 6890 modified (2001–2012) Envirosense 3000i (2012–2015)	Hewlett Packard/Agilent Picarro	GC, FID (CO_2 reduced to CH_4) CRDS
Hoher Sonnblick	CH_4	G2301	Picarro	CRDS
Jungfraujoch	CH_4	G1301 (2010–2011) G2401 (2011.09.16–…)	Picarro Picarro	CRDS
Plateau Rosa	CH_4	Venus 301	NIRA	GC, FID
Zugspitze/Schneefernerhaus	CH_4	HP 6890 modified (2001–2012) Envirosense 3000i (2012–2015)	Hewlett Packard/Agilent; Picarro	GC, FID (CH_4) CRDS

by their exposed position in the high mountain region and the simultaneous efforts to avoid perturbations by local emitters in the immediate surroundings. The four observatories are located along the main Alpine ridge from southwest to northeast whereby the main investigation area (bordered by the perimeters of the Alpine convention) as well as the Alpine foothills and their surroundings are well covered (see Fig. 1). The thereby ensured cover of the Alps as focus region for the CO_2 and CH_4 characterisation of the greater Central European region accompanied by the before outlined highest data quality level and the far-reaching representativity of the measurements offer an ideal data basis in order to reach the project objective.

Methodology

Climate gas data processing

The time series of CO_2 and CH_4 from the atmospheric measurements at the four high-alpine observatories mentioned above form the basis for the characterisation of the regional CO_2 and CH_4 budget of the Alpine region as a focus area of the Central European investigation region. In order to receive statements concerning the origin of the measured CO_2 or CH_4 concentration levels, the high-precision CO_2 and CH_4 measurement series recorded at these four observatories have to be combined with the synoptic transport situations prevailing at the respective measuring time on the individual stations. This requires, first of all, the performance of a climate gas-specific filtering of the CO_2 and CH_4 data for long-term trend and seasonality. The thereby achieved explicit concentration on the short-term varying component in the measurements ensures the exclusive reproduction of the influence of emission strength from sources and relative sinks in conjunction with the meteorology in the following work steps. For this specific filtering, the well-established and frequently applied procedure of Cleveland et al. [12] is used. Their so-called seasonal-trend decomposition procedure based on loess consists of a sequence of applications of the loess smoother. The filtering procedure decomposes a seasonal time series into three components: first, a trend component representing the low-frequency variation in the data together with nonstationary, long-term changes in level. Second, a seasonal component showing the variation in the data due to the seasonal frequency, which in our case is one cycle per year (here the cycle length of the seasonal component is 4380 since this is the amount of the two-hourly climate gas concentration measurements within one year). And finally, the residual component that is the remaining variation in the data beyond that in the seasonal and trend components (for more details, see Cleveland et al. [12]). Therefore, the result of the double filtering reflects the short-term varying proportion of the data (4th line in Fig. 2) without the influence of the long-term, anthropogenic climatic change signal (3rd line in Fig. 2) and the seasonality of the biogenic carbon cycle (2nd line in Fig. 2). These residuals are extracted specifically by station and climate gas. Together with the reconstructed, atmospheric transport conditions of the air masses recorded at the time of the measurements, they constitute the starting point for the estimation of the CO_2 and CH_4 budgets of the Alpine region on basis of the measurement time series.

Backward trajectories

In the present study, four-dimensional back trajectories (three space dimensions plus time) are retrieved using the Lagrangian particle dispersion model (LPDM) FLEXPART. The LPDM FLEXPART further developed at the Norwegian meteorological service simulates the

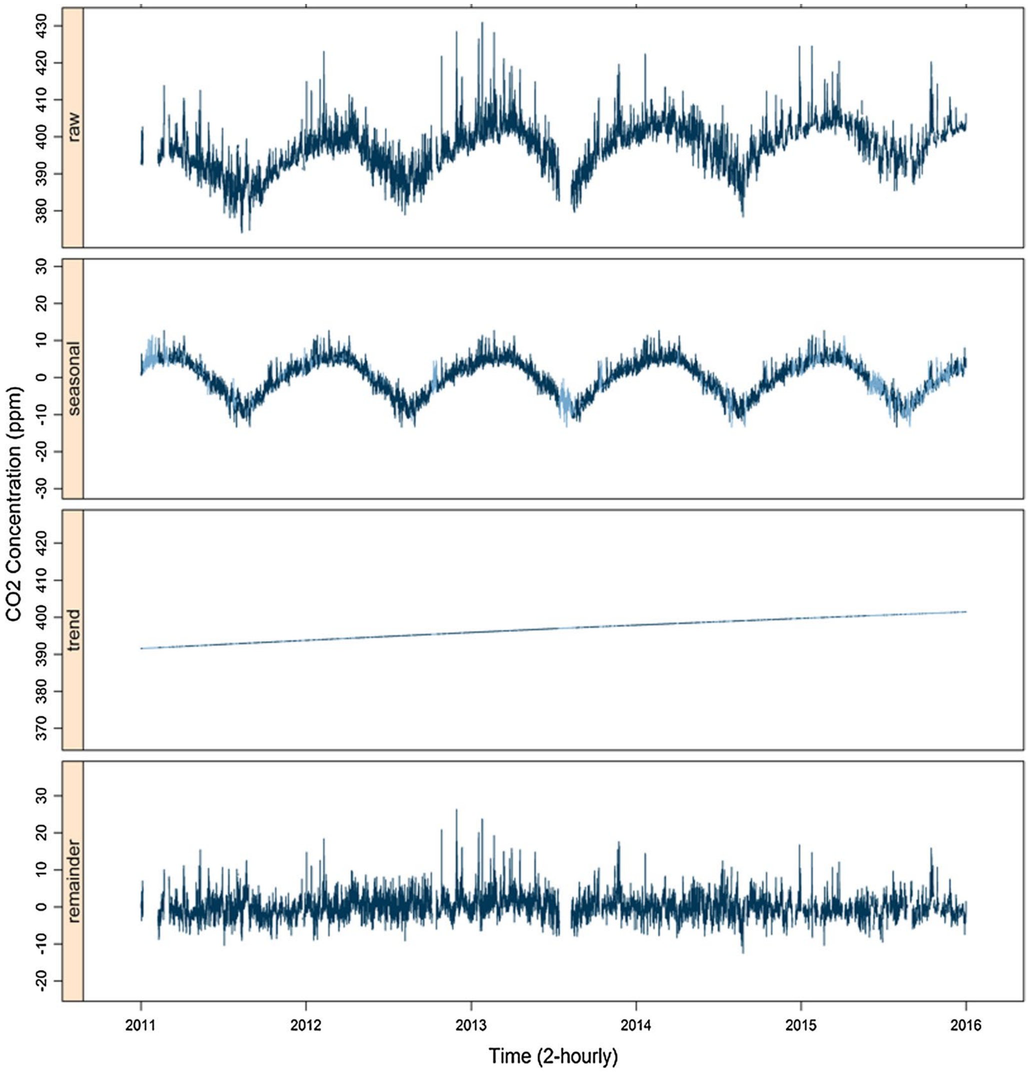

Fig. 2 Seasonality (2nd line) and trend (3rd line) adjustment of the CO_2 concentrations (measured 2011–2015 at Schneefernerhaus) based on the approach of Cleveland et al. [12] and Hafen [15] for the explicit consideration of the transient weather- and emission strength-related components (4th line) within the original CO_2 measurements (1st line)

atmospheric transport of small air volumes on the meso- and large-scale under consideration of diffusion, convection, turbulence as well as dry and wet deposition [30]. For dispersion modelling of pollutants, FLEXPART traces the propagation of gases from a known source forward in time. In its backward mode, however, it serves the allocation of source regions for a certain receptor. The meteorological driving fields of a numerical weather prediction model form the basis of the particle dispersion simulations and thus also its centroid tracks. Since the model of the European Centre of Medium-Range Weather Forecasts (ECMWF) has established itself as the most widely used input base in the dispersion modelling for European study areas, FLEXPART is operated with the meteorological fields of the ECMWF ERA-Interim model (inter alia: [5, 6, 16, 30]). The limited resolution of the ECMWF reanalysis fields as well as of the LPDM (especially considering the horizontal dimension) in combination with the complex terrain in the investigated Alpine domain pose a challenge for any model to accurately describe the transport processes [14, 28]. In order to meet this challenge and in particular the practical problem concerning the differences between the model surface altitude and the real site altitudes, we follow previous experiences from sensitivity analysis with different release heights [5, 6, 16]. These studies show consistently that it works best (independent of time of day) for mountaintop sites to release particles at a medium height between the model surface and the site altitude. From the calculated site-specific release heights in such a way, ten thousand air volumes with the tracer characteristics of the climate gases released every 2 h at the respective receptor observatory are tracked back more than 10 days. The positions of the dispersing particles are stored with a time step of 2 h. The over 17,500 10-day particle dispersion calculations for every year of the investigation period (2011–2015) form the basis for the source contribution studies of the climate-relevant gases together with the CO_2 or CH_4 concentrations measured at the respective release time.

Before combining the FLEXPART results and the climate gas data for analysis, the uncertainties associated with the dispersion simulations such as the limited resolution of the meteorological ECMWF fields and the parameterisations of the LPDM itself are considered by an intermediate pre-processing step. To take the limited reliability of the dispersion modelling into account and to reduce these intrinsic model uncertainties, the backward simulations of the particle dispersions are aggregated to their centroid tracks, which results in the cancelling-out of errors. On the assumption that the uncertainties are equally distributed, the coordinates of the centroid pathways represent the average and thus the least erroneous transport positions of the particle tracking (visually checked for a test sample of FLEXPART particle dispersions with positive outcome).

The site-specific footprints of all backward centroid tracks from the respective receptor observatories of Schneefernerhaus, Jungfraujoch, Sonnblick and Plateau Rosa over the years 2011–2015 bring out the strongly different catchment areas of the four high-alpine sites (see Fig. 3). While the fields of view of the observatories Jungfraujoch and Plateau Rosa are primarily extending on the western or southwestern areas of the Alps and the Alpine foothills, the Environment Research Station Schneefernerhaus covers the northern to north-western regions of the study area that is finally completed from south to northeast by the catchment area of the Sonnblick observatory. If the centroid paths of the particle dispersion simulations of all four receptor sites are combined, these footprints complete each other and form a common catchment area which includes the course of the Alpine ridge and thereby captures the air masses very frequented over the Alpine study region (see Fig. 4).

Concentration weighted trajectory fields

A complex and frequently applied representant from the family of the trajectory-based receptor models are the so-called concentration weighted trajectory fields (CWT) introduced for the first time by Seibert et al. [29] and further developed since. For the calculation of the CWT, the complete study area is subdivided into grid cells. The contribution of every grid cell to the measured climate gas concentrations at the receptor observatories, averaged over the respective study period, arises from the combination of the paths of atmospheric air masses arriving at the measurement sites in the form of backward trajectories with the climate gas concentrations measured at the same time. The formula to calculate such a contribution [2] considers the length of stay of the air parcels over geographic regions prior to their arrival at the receptor site just as the measured concentration level of the climate gas. To avoid mistakes on account of the reduced reliability of the value for grid cells with low intensity of trajectories passing by, the grid cells are weighted according to their frequency of trajectories crossing. This minimises the effect of few trajectory coordinates in individual grid cells and ensures that the increased uncertainties of areas less frequented are taken into consideration [23]. Seibert et al. [29] computed for each grid cell of this domain the mean concentration of the investigated species as follows:

$$\overline{C}_{ij} = \frac{1}{\sum_{k=1}^{N} \tau_{ijk}} \sum_{k=1}^{N} c_k \tau_{ijk}$$

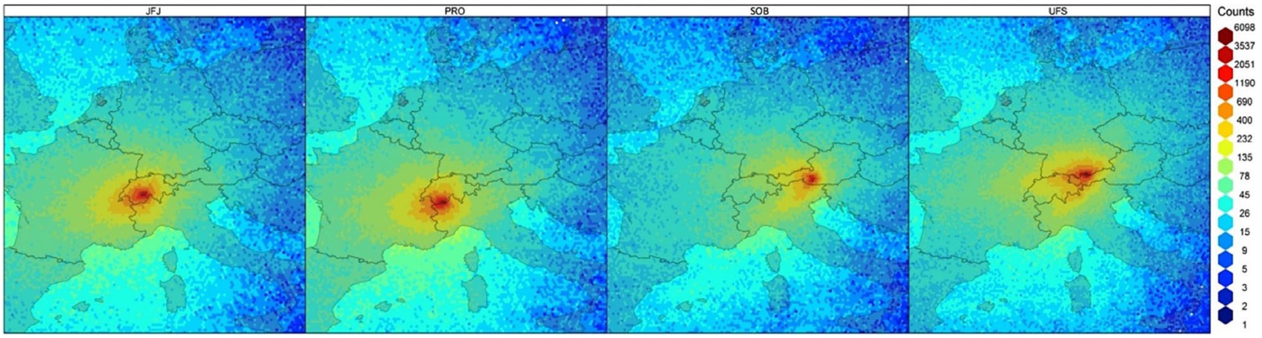

Fig. 3 Footprints of the centroid pathways from the site-specific FLEXPART particle dispersion modelling for the receptor observatories Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS), respectively, visualising the frequentation of the 0.2 × 0.2° grid cells within the individual catchment area by the backward trajectories during the study period 2011–2015

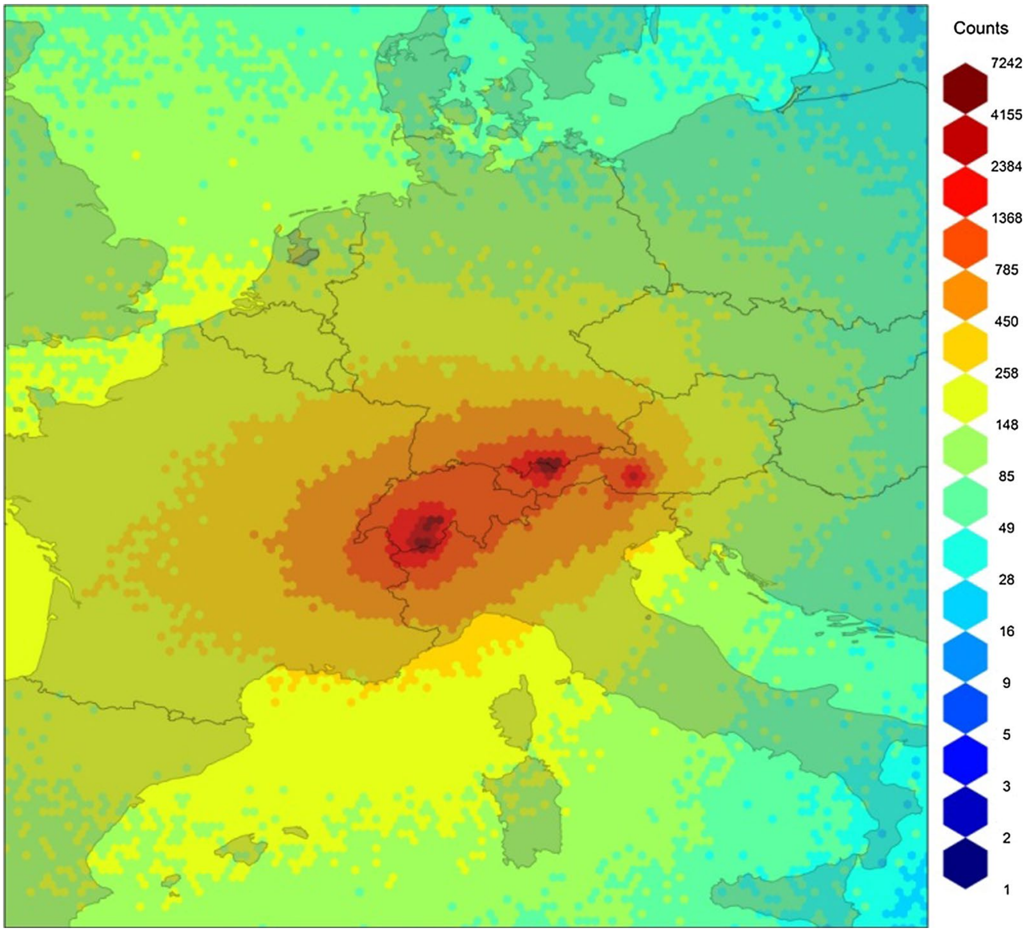

Fig. 4 Combined footprint of the centroid pathways from the site-specific FLEXPART particle dispersion modelling for the receptor observatories Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS), respectively, visualising the frequentation of the 0.2 × 0.2° grid cells within the extended catchment area by the backward trajectories during the study period 2011–2015

where i and j are the horizontal indices of the grid, k the index of trajectory, N the total number of trajectories used in the analysis, c_k the pollutant concentration measured upon arrival of trajectory k and τ_{ijk} the residence time of trajectory k in grid cell (i, j). A high value of $\overline{C}_{ij}$ means that air parcels passing over cell (i, j) would, on average, cause high concentration levels at the receptor sites. On the other hand, a negative value means that the grid cell has on average a concentration lowering influence on the measurements at the receptor sites. According to this formula, the CWT approach is even able to distinguish between moderate sources or sinks and intense ones [7].

Emitting or absorbing grid cells can correspondingly be recognised by high or low values and reveal in their entirety a map that identifies potential source and sink regions with influence on the measurements at the receptor stations with high spatial resolution [9, 17]. Such CWT maps act as reliable indicators for the identification of regions with positive or negative effects on the climate gas concentrations measured at the receptor sites and represent the areas relevant for the measurements precisely, as the comparison with known emission sources has pointed out [2, 8].

Calculation of the CO_2 and CH_4 budgets of the Alps

For the detection of relevant sources and sinks with influence on the CO_2 concentrations of the years 2011–2015 as well as the temporal variability of the emitters and absorbers recorded at the Alpine observatories (UFS, JFJ, SOB and PRO), the climate gas time series of the four measurement stations are first of all subjected to the adjustment for seasonality and long-term trend derived by Cleveland et al. [12]. This ensures that the results of the analyses are distorted neither by the climate change signal nor by the annual cycle, but refer exclusively to the short-term component in the measurement time series that depends on weather and emission strength of the sources or relative sinks.

Parallel to this statistical processing of the measurement data, the calculation of the 10-day centroid tracks derived from the backward particle dispersion simulations with FLEXPART is carried out individually for each of the four receptor sites, but with analogue settings of the model parameters. Altogether, over 85,000 FLEXPART particle dispersion runs have been conducted for this study and then been related to the CO_2 concentrations of the receptor observatories recorded at the same time and subjected to the CWT analysis.

The examination of the CO_2 budget based on atmospheric measurement time series represents a special challenge due to the variety of existing sources and sinks of carbon dioxide whose atmospheric concentration is continuously modified by biogenic as well as anthropogenic emitters and absorbers. If a model performs this complex task plausibly, the relatively simple transfer to other climate gases of similar characteristics, such as methane, is obvious to improve the knowledge progress. For this reason, the model is extended to the examination of the CH_4 budget for the Alpine region whereby the studied region as well as the examination time period and the methodical approach are kept in analogy to the previous CO_2 budget characterisation. The only necessary modification is the consideration of the CH_4 measurement time series instead of the CO_2 data of the observatories.

As done for the CO_2 time series, the CH_4 measured data of the four high-alpine sites are first of all subjected to the adjustment for seasonality and long-term trend [12]. Exclusively the residuals of the measurements which reflect the weather and emission intensities are analysed in the following to illustrate neither the impact of climate change nor biogenic seasonal cycle. For the analyses of the Alpine CH_4 budget, the seasonally and trend-adjusted CH_4 time series of the four stations are set in relation to the site-specific centroid tracks of the FLEXPART particle dispersions for the years 2011–2015. The combination of the residuals of the measurement time series and the backward trajectories is again expressed in site-specific CWT analyses.

Results and discussion

Characterisation of the Alpine CO_2 budget

The result of the combination of centroid tracks derived from the site-specific FLEXPART simulations and the CO_2 concentrations measured there to the respective time of arrival of the backward trajectories after their adjustment for seasonality and long-term trend is shown in Fig. 5 in the form of CWT maps.

These maps for the four receptor observatories over the complete study period 2011–2015 point out that particularly regions of Eastern Europe as well as Central Europe north of the Alps are responsible for high CO_2 concentrations at Schneefernerhaus and Sonnblick. In contrast, high CO_2 measurements at the sites Jungfraujoch and Plateau Rosa can be traced back primarily to the impact from regions south of the Alps. At this, the height difference between the measurement sites has notable effects: it causes the 450–830 m lower-lying observatory of the UFS to be influenced by the lower, free troposphere for shorter periods, resulting in a higher average CO_2 concentration of the recorded air masses. The measurements at the three sites JFJ, PRO and SOB located at altitudes over 3000 m on the other hand represent more frequently background concentrations of carbon dioxide from the well-mixed free troposphere and are not influenced by the contributions of relative sinks or sources within the immediate surroundings. Altogether, both the altitude and the position of the measurement sites within the Alpine core study region are reflected in the site-specific CWT maps, which clearly show different focus regions of the individual footprints, where sources and relative

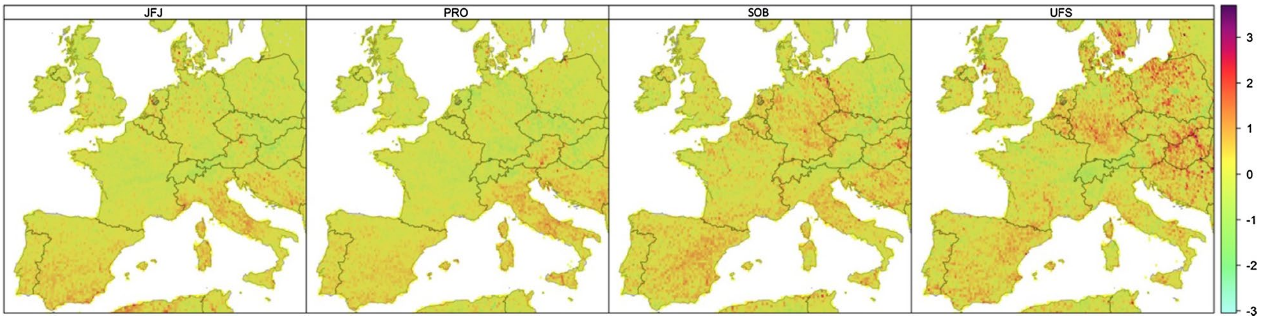

Fig. 5 Concentration weighted trajectory fields quantifying the site-specific influence of source and relative sink areas to the de-seasonalised and de-trended CO_2 concentrations (in ppm) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS), respectively, over the entire study period 2011–2015

sinks strongly influence the measurements at the receptors (more intensive colouring in Fig. 5).

The different coverage of the probed area corresponding to the locations of the measurement sites indicates the significance and need for a broad scope of the database to time series of more than just one observatory for studies like ours. As found by Kaiser et al. [19], the ability of the model to reliably identify sources and sinks is directly linked to the number of station data taken into account. Consequently, an expansion of the analyses to more CO_2 measurement sites is expected to result in an improved model quality and increased reliability of the results [1, 4]. In particular, the integration of additional measuring stations with (over-) regional representativeness promises more reliable results of the identification of potential source and sink areas, because these measurement time series are hardly influenced locally, and instead detect regional CO_2 as well as CO_2 transported over long distances [36]. For these reasons, only a combination of the catchment areas of at least these four high-alpine sites ensures that the most relevant emitters that influence the climate gas concentrations of the Alpine region are captured.

The result of the cumulative consideration of the CO_2 concentrations and particle dispersion simulations of all four high-alpine observatories in the form of the combined CWT map (see Fig. 6) locates CO_2 emitting regions all around the central Alps with the exception of France in the west which has pulled out of coal mining since the beginning of the 2000s and uses the quite CO_2-neutral nuclear power as main source of energy today. Furthermore, particularly the area around the Alpine main ridge appears as an important large-scale relative CO_2 sink of Central Europe averaged over the

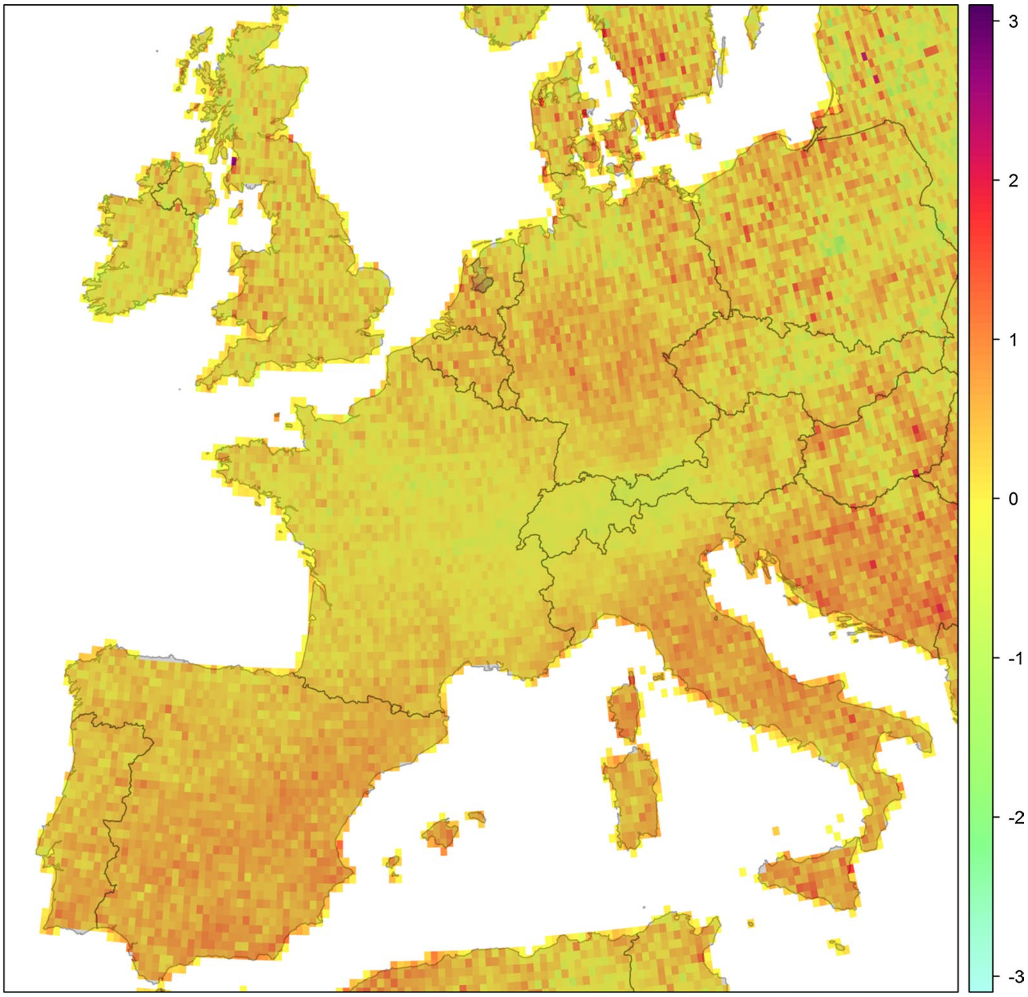

Fig. 6 Combined concentration weighted trajectory field quantifying the influence of source and relative sink areas to the de-seasonalised and de-trended CO_2 concentrations (in ppm) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) over the entire study period 2011–2015

years. Air masses originating from this central region in the midst of the study area caused a significant reduction in the CO_2 concentration levels in the measurements of the years 2011–2015 when recorded at the receptor sites.

The seasonally differentiated maps of the CO_2 contributions from the grid cells of the accumulated catchment area (see Fig. 7) identify single emission hotspots emerging in different seasons. In winter, these are located primarily north and east of the Alps and suggest CO_2 emissions by heating with fossil fuels, whereas during summer CO_2 measurements higher by about two ppm occur mainly during air mass advection from the Mediterranean area southwest of the Alps as well as from Central Italy. The enhancement of Alpine concentrations of carbon dioxide caused by emissions of burning fossil fuels, which amounts to values of up to four ppm in winter, is already ascertainable in fall—though less strong—and can in this season be attributed to eastern European regions located further inland. These most dominant emission regions include parts of the northeast of Germany and particularly wide areas of (West-)Poland and Eastern Europe. Given the fact that the biggest brown coal-mining areas of Europe are located in these regions, this indicates the considerable impact of the brown coal

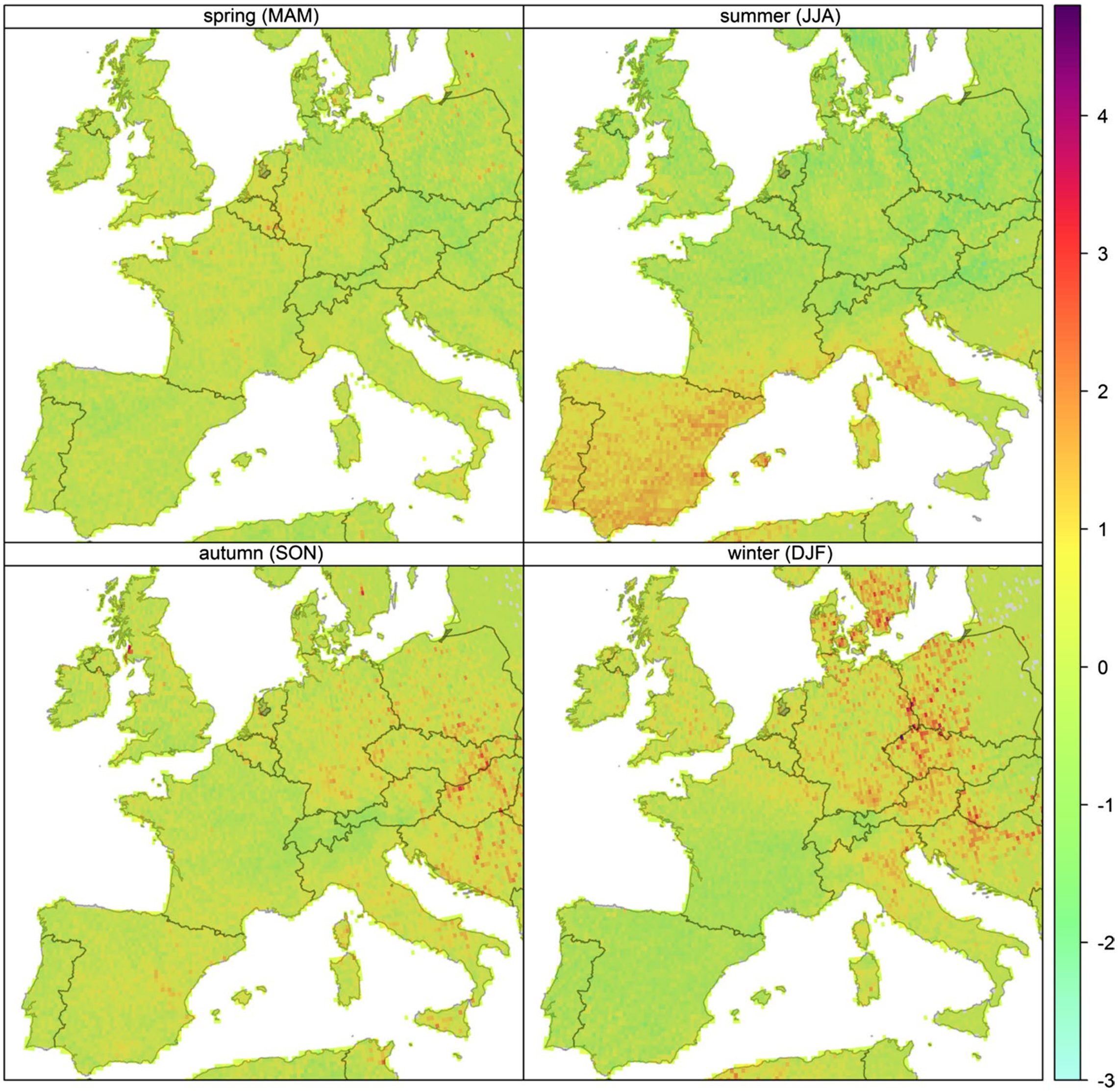

Fig. 7 Concentration weighted trajectory fields quantifying the seasonal influence of source and relative sink areas to the de-seasonalised and de-trended CO_2 concentrations (in ppm) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) over the entire study period 2011–2015

emissions on CO_2 measurements, even at the high-alpine observatories situated in over 500 km straight-line distance at the top of Europe's highest mountains. The increases in the carbon dioxide level detected at the high-alpine sites during summertime stem from the north-west Mediterranean region where at this season often heat-related fires spark off. The CO_2 released in these may well be responsible for values of CO_2 higher by up to three ppm on average at the receptor sites, when air mass transport originates from this area during summer. Spring, in turn, shows some less contributing CO_2 emission hotspots in highly populated areas of West Germany (Rhine-Ruhr area), Belgium and the Netherlands. The vehicular and power plant emissions from these regions are clearly displayed only during this season when private heating and wildfires do not play any role suggesting that the latter are the major sources for enhanced CO_2 concentrations measured at the Alpine sites. Other large European metropolitan areas such as Paris or London may be located too far away from the four high-alpine observatories for a significant detection so that their impact thins out over the long distance (and especially over the Atlantic Ocean in the case of London). During all seasons, the central study area around the Alpine main ridge is classified as a significant relative carbon dioxide sink whereupon the Alpine core region shows the largest negative influence on the CO_2 concentrations of Central Europe over all seasons.

Altogether, these seasonally differentiated analyses of the Alpine CO_2 budget on the basis of the combination of the measurement time series of the stations UFS, JFJ, PRO and SOB draw a plausible picture of the seasonally relevant CO_2 emitters and absorbers near enough to the Alpine receptor sites and underline at the same time the relevance of seasonal differentiations of the CO_2 source contribution studies. The seasonal variations of the emitters and absorbers, very distinctive in particular for carbon dioxide, appear only in seasonally differentiated CWT analyses as performed here. These allow in their entirety a conclusive characterisation of the relevant regions of Europe with influence on the Alpine CO_2 concentrations.

In the CWT maps calculated separately for each of the 5 years of the study period (see Fig. 8), the high CO_2 emissions due to the severe winter 2013 with temperatures down to −20 °C clearly stick out. Only after lots of snow during March, that according to meteorological categorisation already counts to one of the spring months, this came to an end. The methodology visualises the intensified and longer-lasting emissions from heating with fossil fuels caused by these weather conditions reliably for the main key region Central Europe. Likewise, the method also succeeds in representing the following particularly mild winter 2014 with its weather-related much lower CO_2 emissions from the reduced incineration of fossil sources of energy. The reliable reconstruction of the year-to-year variations in the CO_2 emissions using the year-wise CWT analyses stress the effectiveness of the methodology to trace back the short-term varying components within the measurements of the high-alpine observatories reflecting the influence of weather and emission strength to their source regions. In summary, this certifies the ability of the method used here to identify both seasonal and year-wise deviations of the CO_2 emissions from areas with influence on the CO_2 budget of the Alpine study region.

Characterisation of the Alpine CH_4 budget

As already seen in the source contribution studies for the CO_2 concentrations of the stations, site-specific focuses within the Alpine investigation area also appear in the concentration weighted trajectory fields for the CH_4 data (see Fig. 9). While the observatories JFJ and PRO are identifying the methane source regions north and

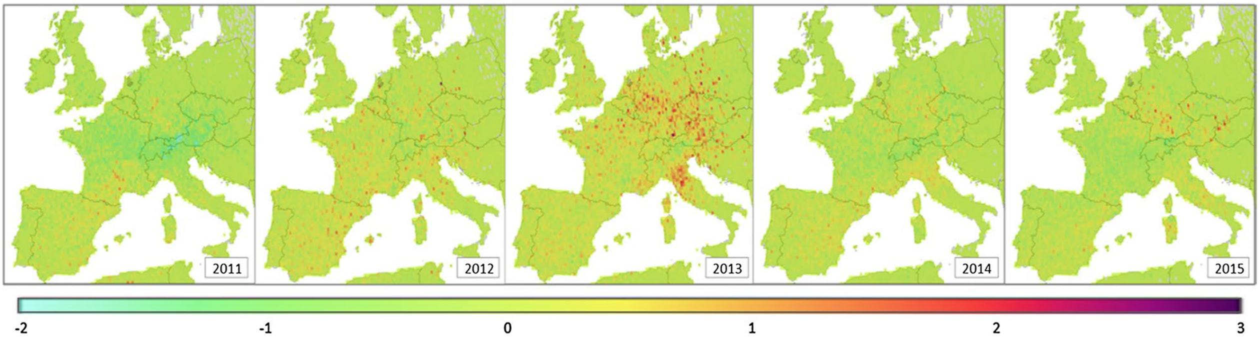

Fig. 8 Concentration weighted trajectory fields quantifying the yearly influence of source and relative sink areas to the de-seasonalised and de-trended CO_2 concentrations (in ppm) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) over the entire study period 2011–2015

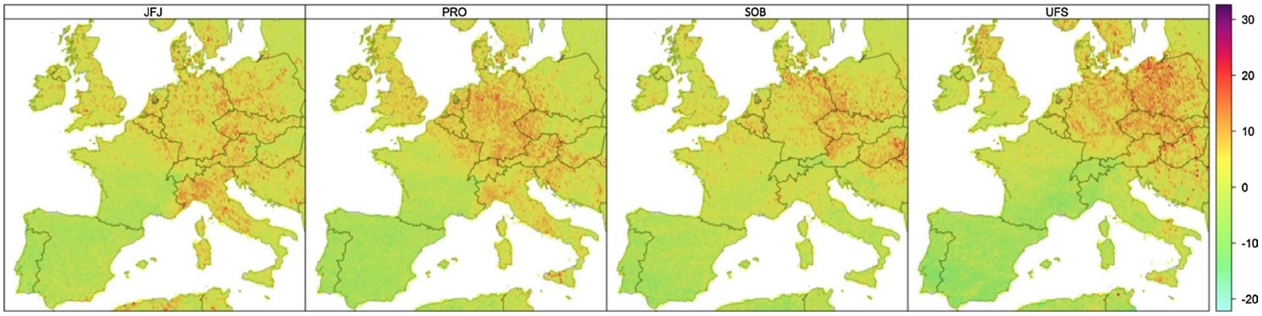

Fig. 9 Concentration weighted trajectory fields quantifying the site-specific influence of source and relative sink areas to the de-seasonalised and de-trended CH_4 concentrations (in ppb) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS), respectively, over the entire study period 2011–2015

(even more) south of the Alps, emitting grid cells in the north and east of Central Europe are detected primarily at Sonnblick and Schneefernerhaus. Hotspot regions—even on the small scale—are localised correspondingly in several site-specific maps, what increases the reliability of the cumulative source detection. To all four site-specific CWT maps, the consistent classification of the southwest of Europe as a large-scale relative CH_4 sink is common, regardless of their individual footprint. Thus, air masses of the Iberian Peninsula, when recorded at the Alpine observatories, caused mean reductions in the CH_4 concentrations measured there of about ten ppb averaged over the whole 5-year investigation period. On the other hand, air mass transport particularly from the northeast of Central Europe is accompanied by increased CH_4 concentration levels of up to 20 ppb on their arrival and recording at the high-alpine sites during the years 2011–2015.

The site-specific CWT maps already give a picture of the division of the catchment area in two parts with a southwestern relative CH_4 sink region on the one hand (Portugal, Spain and the south of France) and CH_4 emitters (from England to Italy with main focuses in the northeast of Central Europe: Germany, Denmark, eastern Austria, Poland, Czech Republic, Hungary) on the other hand. This phenomenon of a clear regional split is confirmed by the comprehensive plot incorporating the calculations from all four stations (see Fig. 10). This CWT map of the combination of all station data and backward trajectories for the whole period 2011–2015 manifests the strong northeast-southwest gradient in the impact to the Alpine CH_4 concentrations according to which the seasonally and trend-adjusted measured values increase by 15 to 20 ppb during advection of air masses particularly from the northeast of Central Europe, while air masses from the southwestern direction cause a negative influence on the measurements in form of a reduction of 15 ppb CH_4 on average. The only exception to this northeast-southwest distinction is the Alpine region itself whose effect as a relative CH_4 sink considerably emerges despite the surrounding source regions. One can therefore generally constitute that the Alpine core region represents a significant relative sink for both examined climate gasses (CO_2 and CH_4) in the midst of an emission-intensive Europe and thus merits a special protection status also from the climate protection perspective.

The northeast-southwestern division in CH_4 sources and relative sinks keeps its validity also for the seasonal consideration of the influence from the grid cells on the Alpine CH_4 concentrations (see Fig. 11). In this merging of the site-specific calculated seasonal concentration weighted trajectory fields, the Iberian Peninsula appears in all seasons as an extensive area of origin for methane poor air masses in accordance with the previous results. A plausible explanation for this is the lack of wetlands in the characteristically dry Mediterranean area. Wetlands are the most important natural methane source and react very sensitively to climate changes and weather anomalies like higher precipitations and temperatures. Methane emissions from wetlands are triggered strongly by the water supply, because higher water levels on account of high amounts of precipitation promote anaerobic conditions favouring methane formation. Decomposition processes of methane, in turn, can be favoured by higher temperatures. The anthropogenic sources like rice fields and the burning of biomass are also subject to the influence of these climatic factors, so that warmth and humidity have in general great effects on the intensity of the CH_4 emissions. This influence can be found again in the seasonal maps, not only regarding the southwestern European dry regions and their air masses poor in CH_4, but is also in consideration of the high CH_4 emissions from regions with wide, natural or artificial (in particular

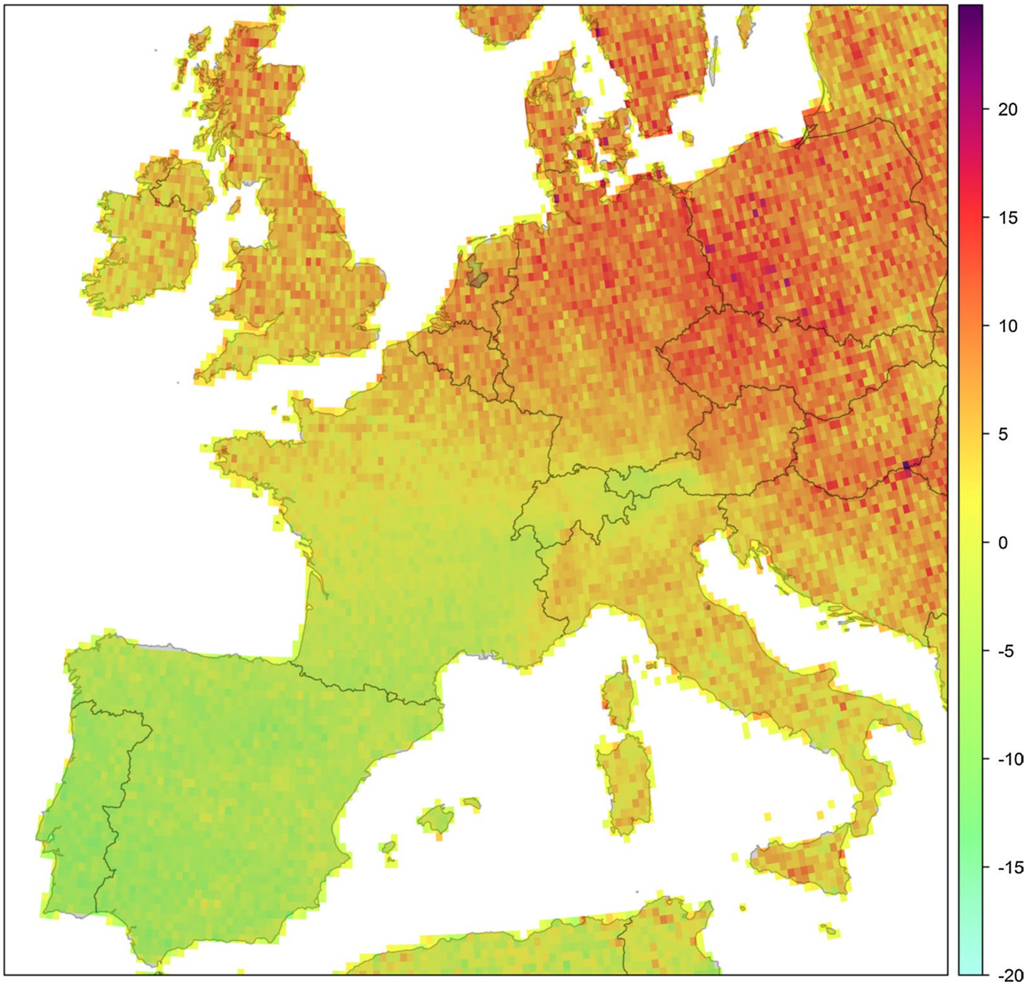

Fig. 10 Combined concentration weighted trajectory field quantifying the influence of source and relative sink areas to the de-seasonalised and de-trended CH_4 concentrations (in ppb) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) over the entire study period 2011–2015

flooded, former mining areas) wetlands as for example in Poland. The concentration increasing influence of higher temperatures gets visible in the seasonal CWT maps in the form of more intensive CH_4 emissions in summer and fall. In the winter season, the northernmost source regions (east of Poland, Slovakia, Hungary and Lithuania) for high CH_4 concentrations measured at the Alpine sites disappear, since wetlands there are covered by snow and/or frozen and don't emit anything. The emitting regions further west during winter may represent wood burning for heating purposes as also seen in the analogous CWT maps for CO_2. Nevertheless, the CH_4 emissions from fires are many times lower than the amount of CO_2 that is emitted at the same time [21]. That's also the reason why wildfires in the Mediterranean region aren't visible emission hotspots in the seasonal CH_4 CWT maps, but an important source in the equivalent figure for CO_2.

The basic phenomena of the detection of source and sink regions with southwestern Europe known by negative CH_4 contributions and emission-intensive regions in the north(west), the east and the south of Europe are also evident in view of the annual analysis of the influence on the Alpine CH_4 concentrations (see Fig. 12). In consistency with the previous results, the central Alpine region finds itself in all five yearly aggregated CWT maps always as an area of negative contributions to the measured CH_4 residuals. The differences between the years can again be explained by the climatic factors precipitation and temperature. Accordingly, years as well as regions with high precipitation sums, mostly in connection with warm summers and mild wet winters, effect high annual contributions to the CH_4 concentrations of the Alpine region.

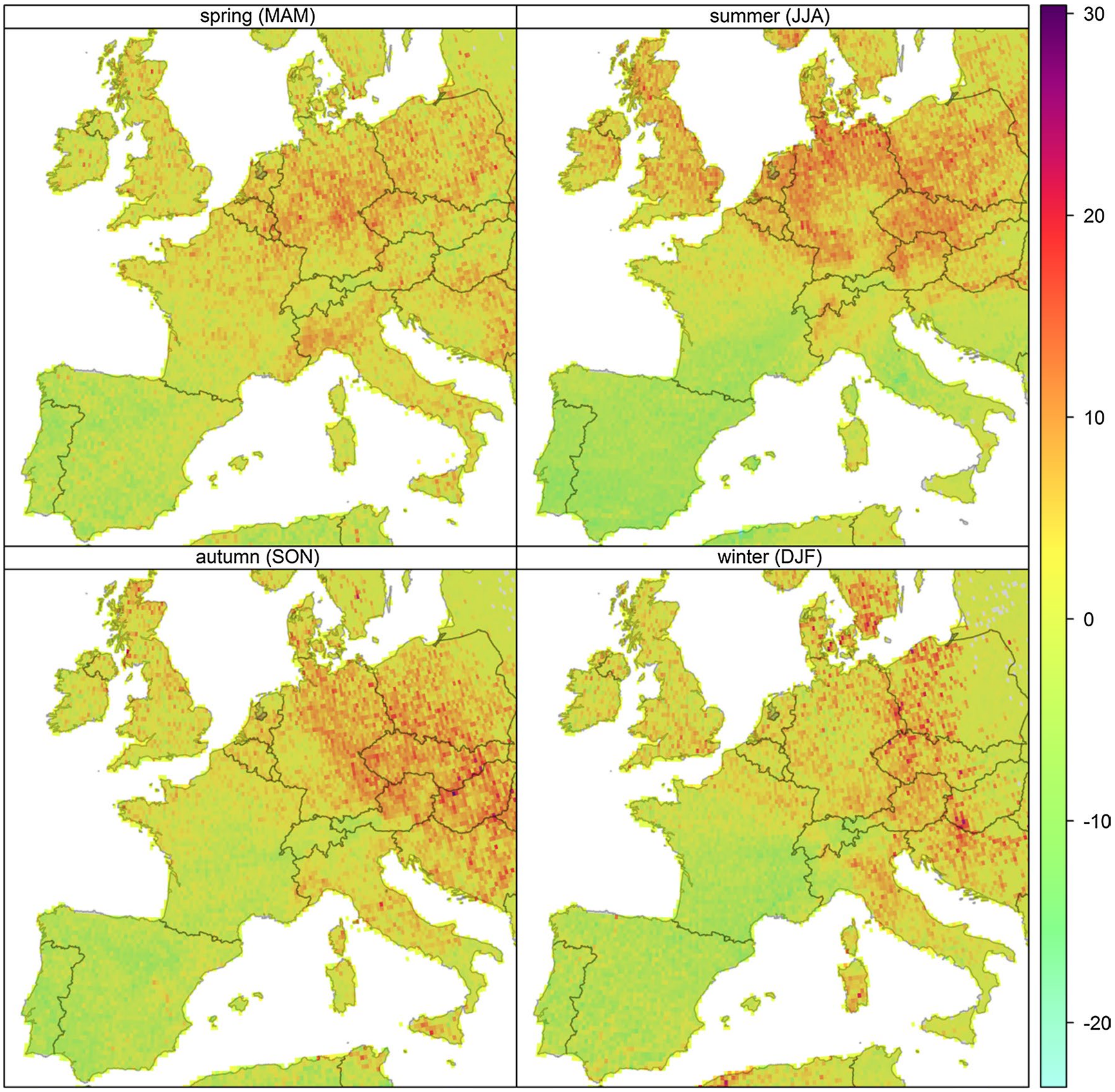

Fig. 11 Concentration weighted trajectory fields quantifying the seasonal influence of source and relative sink areas to the de-seasonalised and de-trended CH_4 concentrations (in ppb) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) over the entire study period 2011–2015

The range of the annual differences in the contributions can approximately be compared to that of the seasonal contributions and contains values of −20 to +20 ppb averaged over the 5-year study period.

Uncertainty assessments

In comparable studies on the combination of measurement time series with trajectory analyses, Reimann et al. [24] find that this has the potential to conclude on European emission quantities and to act as independent tool for the verification of anthropogenic trace gas emissions as they are determined in international contracts like UNFCCC [35]. For an assessment to what extent also the method applied here and the results obtained with it allow for reliable conclusions on emitters and relative sinks combined with their temporal variability and in addition to a logical interpretation of the results, a quantification of the uncertainties connected with the outcomes is required.

In principle, the reliability of a grid cell value is directly connected to the amount of trajectories crossing the same grid cell. Because the larger the number of single trajectory coordinates is within a grid cell, the higher is the certainty that the value calculated for that grid

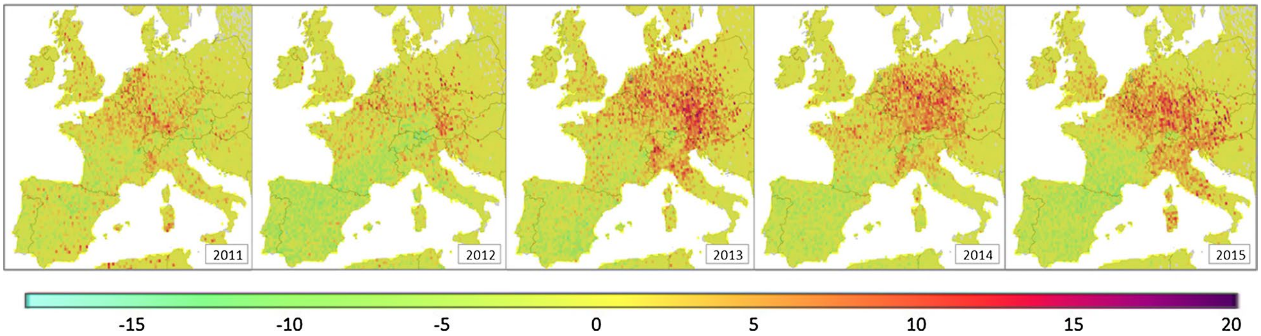

Fig. 12 Concentration weighted trajectory fields quantifying the yearly influence of source and relative sink areas to the de-seasonalised and de-trended CH_4 concentrations (in ppb) at the high-alpine receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) over the entire study period 2011–2015

cell corresponds to reality and doesn't reflect a random extreme value or exception. Thus, with increasing frequency of trajectories, the reliability of the calculated contribution from the grid cells of the investigation area to the climate gas measurements at the Alpine observatories rises accordingly. As we have four high-alpine sites as receptors of the particle dispersion modelling with FLEXPART, the grid cells of the strongest frequentation, and therefore with the highest reliability, are in close vicinity to these as well as in the central area of the Alpine investigation region spanned by the four stations, which corresponds approximately to the perimeters of the Alpine convention (see Fig. 4). Due to the prevailing westerly wind direction over the analysis area by its location within the west wind zone, the further extension of the footprint representing the frequentation of the individual grid cells by the backward trajectories is not symmetrical in all directions, but shifted to the west. Altogether, more than 5% of the trajectory coordinates of the particle dispersion calculations during the 5-year investigation period 2011–2015 lie within the Alpine centre of the study region including the Alpine foreland. Therefore, a high reliability and explanatory power can be attributed to the results of this focus region.

Besides the frequency of trajectories, the intensity of the contact with the planetary boundary layer close to ground is another important criterion to estimate the reliability of the project results. The stronger the boundary layer contact within a grid cell is, the higher is the probability that the basic assumption underlying the model of the absorption and the effective transportation of the changes of the atmospheric trace gases is realised while air passes a grid cell with sources or sinks. In analogy to the footprint, the highest values within the representation of the average proportional boundary layer contact derived from the backward trajectories for each grid cell during the analysed years 2011–2015 are also found near the receptor observatories where trajectories model intrinsically arrive in close proximity to the ground level. Over the further Alpine region and its foreland, the contact with the boundary layer during the study period consists of at least 20% on and also doesn't drop below mean average values of ten percentage over the remaining European continent except for the eastern edge area (see Fig. 13). These comparatively high values underline the plausibility of the exchange process with the near-surface boundary layer assumed in the model, lending (just as the considerations on the footprint) credibility to the results for the core study region of the Alps and the Central European surroundings.

Plausibility checks

Co_2

The model intrinsic estimates of the uncertainties and limitations of the reliability of the results serve, just like comparisons with other models, to judge the model quality. The latter complete the confirmation of the reliability and validity of the model as an external quality control. Because no analogous models in the strict sense that devote themselves to the same scientific issue are available for a comparison with the approach used here, the plausibility checks rely on the results from the inverse modellings of climate gas fluxes and concentrations from the prominent projects Copernicus Atmosphere Monitoring Service (CAMS) (https://atmosphere.copernicus.eu/) and Jena CarboScope (http://www.bgc-jena.mpg.de/CarboScope/). To this end, the fluxes or concentration data of both models are case specifically compiled and compared to our results so that an assessment of the plausibility can be met.

The CAMS CO_2 surface fluxes are estimated with a variational Bayesian inversion system at a resolution of 3.75 × 1.9 degrees (longitude–latitude) and 3-hourly time steps, based on CO_2 mol fraction station records

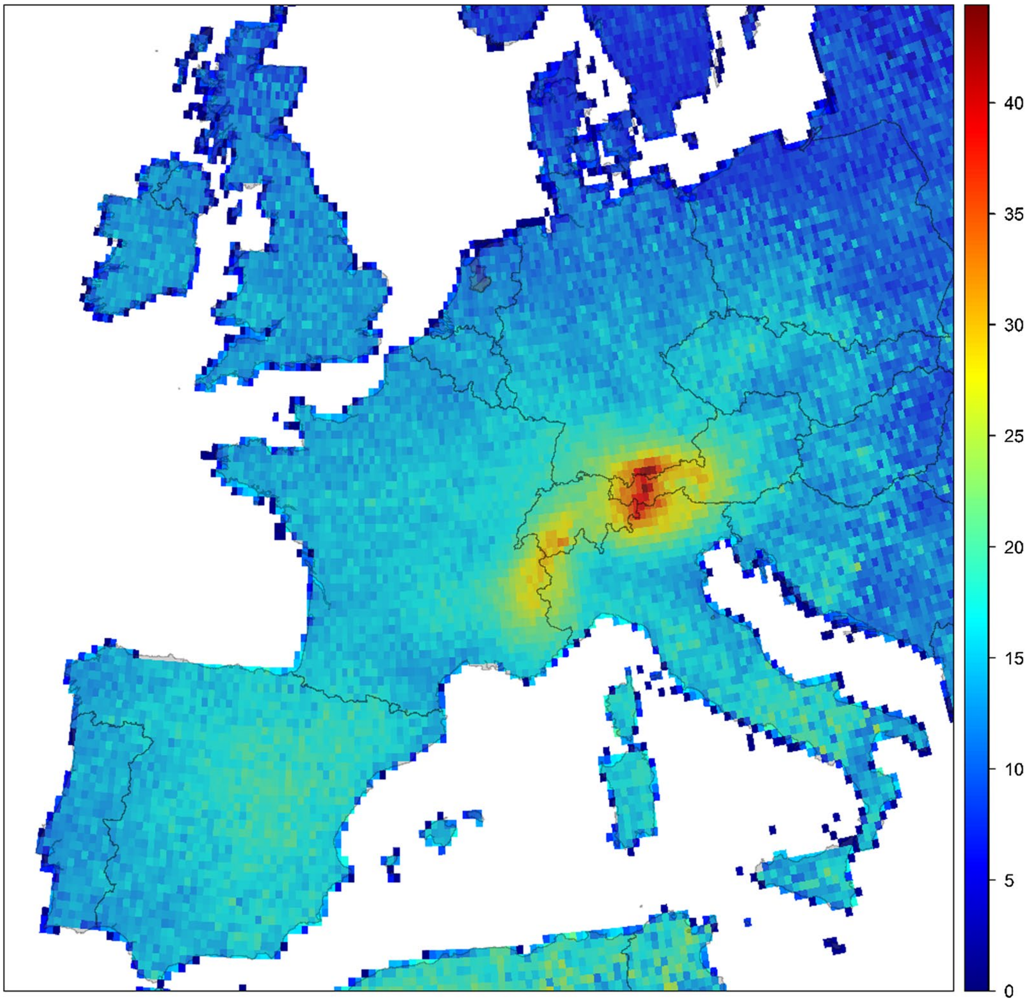

Fig. 13 Mean percentual contact of the centroid tracks out of the FLEXPART particle dispersion modelling from the receptor sites Jungfraujoch (JFJ), Plateau Rosa (PRO), Sonnblick (SOB) and Schneefernerhaus (UFS) with the near-surface planetary boundary layer during the study period 2011–2015 (in %)

from the following large databases including both in situ measurements made by automated quasi-continuous analysers and irregular air samples collected in flasks and later analysed in central facilities:

- The NOAA Earth System Research Laboratory archive (NOAA CCGG),
- The World Data Centre for Greenhouse Gases archive (WDCGG),
- The Reseau Atmospherique de Mesure des Composes a Effet de Serre database (RAMCES)
- The Intergrated Carbon Observation System—Atmospheric Thematic Centre (ICOS-ATC).

Fluxes and mole fractions are linked in the system by the global atmospheric transport model of the Laboratoire de Météorologie Dynamique. More detailed information can be found at Chevallier et al. [10] or Chevallier et al. [11].

For comparison with the project results from Chapter 4.1 for the purpose of plausibility checks, the 3-h intervals of fossil emissions are seasonally aggregated together with the biospheric (posterior) fluxes for the analogous study region of Central Europe with the Alps in the centre over the years 2011–2015. The result is shown in Fig. 14, where negative CO_2 surface fluxes are visualised in blue tones and red tones represent positive CO_2 surface fluxes. In these seasonal maps provided for the comparisons on basis of the CAMS data, it is immediately apparent that the sign in the amount of the combination of biogenic and fossil CO_2 surface fluxes shifts with the seasons. While negative expressions of the CO_2 surface fluxes of over 1 kg of carbon per m^2 and

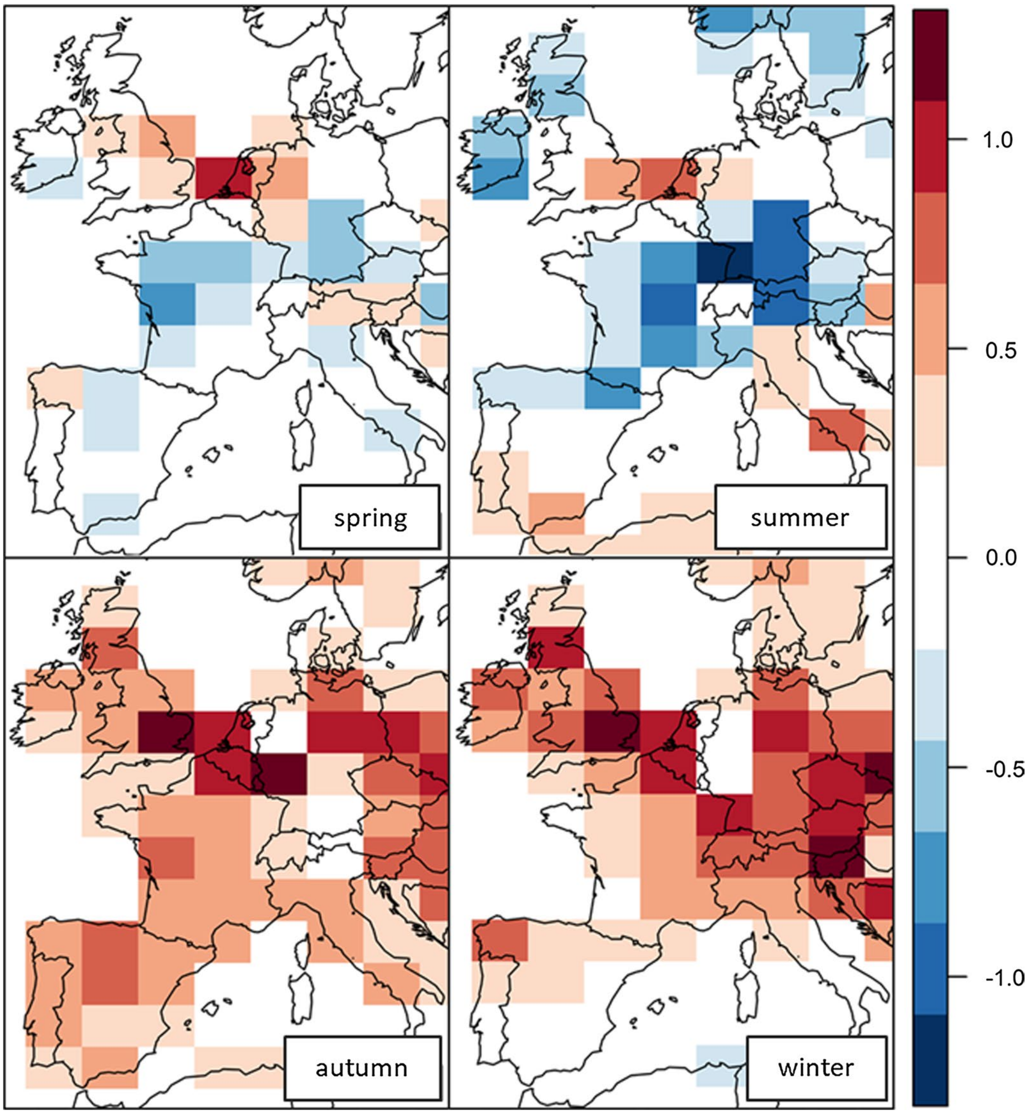

Fig. 14 Seasonally aggregated mean CO_2 surface fluxes of the Copernicus Atmosphere Monitoring Service for fossil and biospheric (posterior) emissions of the years 2011–2015 over Central Europe (in kg carbon/m^2/month)

month are prevailing particularly over large parts of the continent in summer, the positive CO_2 surface fluxes dominate in winter, and—a little more weakly—already in fall over all of Central Europe, with hotspot areas in the east and northwest. The latter emission hotspot is not seen in its intensity in the comparable CWT maps what might be due to the too large distance to the high-alpine receptor observatories. Over this wide distance in connection with the thinning out effect of the Atlantic in the northwest of Central Europe, changes of the climate gas concentration close to ground cannot be detected adequately any more by stations in the Alps and traced back by means of trajectories. At this point, the spatial limitations of the CWT analysis based on the Alpine measurement time series appear (see Chapter 5 and Fig. 13). Nevertheless, our study returns the seasonal variations of the source and relative sink regions of the Central European mainland with the exception of the northwest of Europe during 2011—2015 in high correspondence with the CAMS CO_2 surface fluxes. More detailed, value-based comparisons are not possible due to the very differing spatial resolutions as well as the unequal units of both methodical approaches since surface fluxes and concentration anomalies taken as proxies of emission and deposition fluxes are compared.

In addition, the Jena CarboScope project provides estimates of the CO_2 fluxes between the earth's surface and atmosphere as well as modelled CO_2 concentration fields of different altitudes in global coverage. The latter are calculated using forward simulations of the atmospheric transport model TM3 based on meteorological reanalysis fields. The input data of these simulations are inverse-modelled CO_2 fluxes, which in turn are based on the same transport model in conjunction with observation data. Values between the measuring stations taken into account are estimated by means of interpolation. The resulting CO_2 concentration fields are output in the form of NetCDF files with a resolution of $5 \times 3.75°$ in the unit of ppm. With this, the Jena CarboScope project provides estimates of the surface-atmosphere CO_2 exchange based on atmospheric measurements with a focus on its temporal variations [25, 26].

The near-surface atmospheric CO_2 concentration fields of the atmospheric pressure at mean sea-level of 1013 hPa from the most current version of the Jena CarboScope model runs (s04_v4.1) for the analogous investigation period 2011–2015 and the region of Central Europe with the Alps in the centre are yearly as well as seasonally aggregated for an additional external plausibility check. In the comparison of our results to the yearwise compilation of the near-surface CO_2 concentration fields of the Jena CarboScope project (see Fig. 15), clear common characteristics are recognisable, as for example the localisation of the highest positive CO_2 contributions or concentrations, respectively, over all years in the northern regions of Central Europe as well as their strongest occurrence during the 5-year study period in the year of the intensive winter, 2013. Negative annual means of the CO_2 contributions or concentrations, respectively, appear in the southwest of Central Europe. These regions of positive or negative CO_2 anomalies are returned in the comparable representations not only in their coarse localisation, but in addition also in their scale in very good correspondence of both models. So, both project results show annual amplitudes of approximately five ppm in this comparison. These parallels in spite of the differing model approaches and in particular their disparate resolutions underline once more the reliability of the characterisation achieved in the present analysis of the Alpine CO_2 concentrations with focus on the influence of surrounding source and relative sink regions as well as their temporal variability.

Also in the comparison of the seasonal configurations of both model data (see Figs. 7, 16), fundamental features are found to agree, such as the clear identification of the two regions of positive CO_2 anomalies, the northwestern Mediterranean area in summer as well as the eastern regions of Central Europe in the winter months. The areas of negative CO_2 contributions or concentrations, respectively, are also similarly localised in their seasonal occurrences by both models, whereupon especially in winter, but even already in the transitional seasons the southwest of Central Europe, and in summer in particular the north-eastern areas of the investigation region stick out. Beside the similar localisation of the CO_2 source and relative sink regions in the course of the year, the amplitudes within the single seasons are comparable. Altogether, these parallels found when comparing the seasonally aggregated CO_2 data of both models bring out a substantial similarity of both models in the identification of locations and intensity of the sources and relative sinks with influence on the atmospheric CO_2 concentration as well as their seasonal variability. These agreements despite the distinctive differences in the underlying project objectives and methods again encourage us to positively assess the reliability of the presented study and its underlying methodology.

CH_4

The calculation of the CAMS CH_4 surface fluxes is built up on a four-dimensional, multi-parameter data assimilation system for inverse modelling based on the TM5 atmospheric transport model [3, 22]. Remote sensing

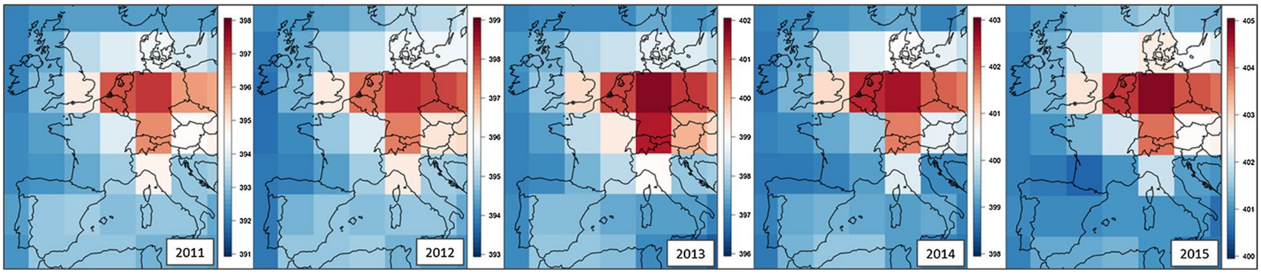

Fig. 15 Yearly mean CO_2 concentrations (in ppm) at the surface over Central Europe aggregated from the Jena CarboScope estimates of the surface-atmosphere CO_2 exchange based on atmospheric measurements (run ID version s04_v4.1, C. Rödenbeck) for the analysed time period 2011–2015

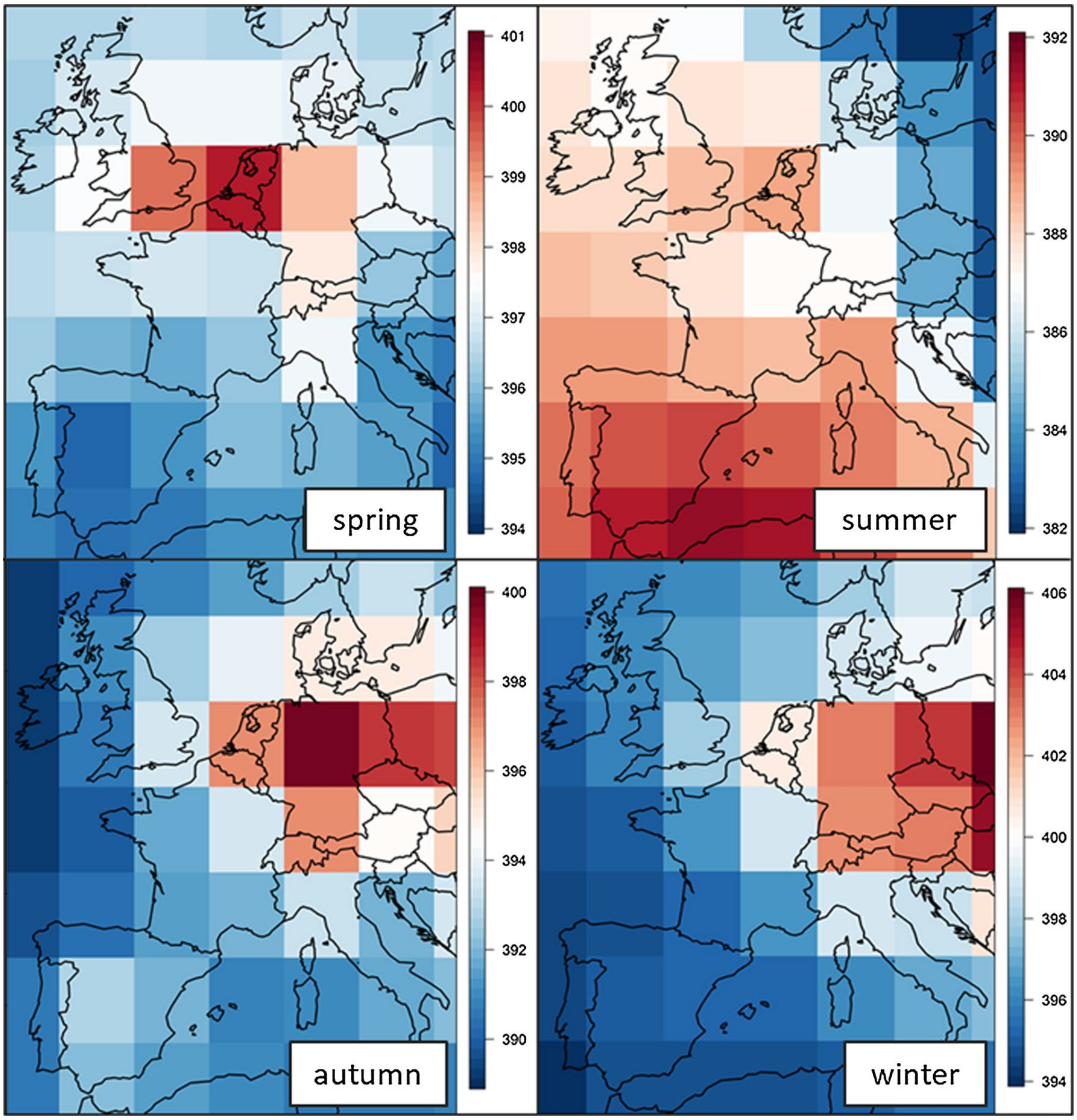

Fig. 16 Seasonal mean CO_2 concentrations (in ppm) at the surface over Central Europe aggregated from the Jena CarboScope estimates of the surface-atmosphere CO_2 exchange based on atmospheric measurements (run ID version s04_v4.1, C. Rödenbeck) for the exemplary year 2011

data from the SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) instrument of the European environmental satellite Envisat as well as high-precision CH_4 measurements of the NOAA network penetrate into the inversion set up. Further details about the model can be found in Segers and Houweling [27].

The complete methane fluxes of CAMS for the study period 2011–2015 have been aggregated seasonally (see Fig. 17) in analogy to the corresponding seasonal CWT maps (see Fig. 11). The comparison of the two plots shows that both models hardly indicate seasonal differences for the methane input of Central Europe and detect, moreover, no significant CH_4 emissions from the Iberian Peninsula (regardless of the season). The latter region is characterised just the same in the CWT approach so that both methods declare correspondingly a negative impact from the north-western Mediterranean area to the Alpine CH_4 measurements. From the more northern regions of Central Europe, in turn, partially very high positive source contributions are detected at all seasons. On this occasion, the main focus in the

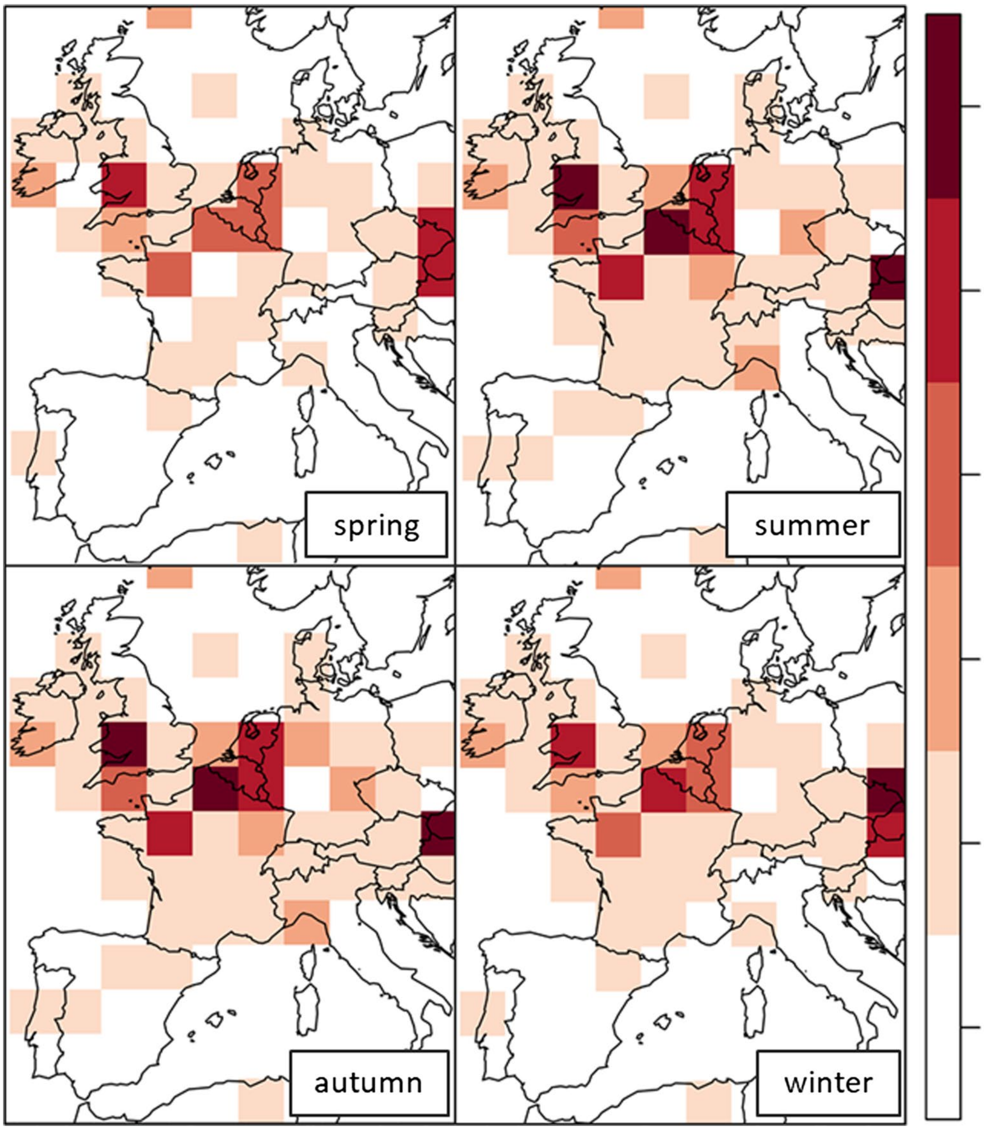

Fig. 17 Seasonally aggregated mean CH_4 surface fluxes of the Copernicus Atmosphere Monitoring Service for the total methane emissions of the years 2011–2015 over Central Europe (in µg $CH_4/m^2/s$)

CAMS CH_4 surface fluxes is found in the northwest, whereas the CWT method localises the methane sources in particular in the north and the east. Thus, the strong northeast-southwest gradient of CH_4 emissions as in the CWT maps cannot be seen in the CAMS data, since the CAMS CH_4 surface fluxes rather point out clear maxima in the highly populated areas (London, Paris, Netherlands and Belgium) which don't appear in the results of the CWT analyses in return. This is due to the catchment area of the CWT analyses being restricted to the Alpine region and its further surroundings as previously seen also in the comparison for the CO_2 emission estimates ("Characterisation of the Alpine CO_2 budget" and "Co_2" sections). This is owed to the position of the four considered Alpine observatories: Schneefernerhaus and Sonnblick still cover well the areas to the east and to the north of the Alps with their footprint, but do not capture the air masses from the far northwest of Europe in sufficient frequency and intensity any more. Here, the thinning effect (strengthened further for sources in England due to the unavoidable crossing of the Atlantic Ocean) demonstrates the limitations of the CWT approach since the impact of emitters located in the northwest of Europe is far too distant to still be captured reliably by measurements at the Alpine observatories. Furthermore, westerly winds are typically accompanied by a lower degree of stability within the vertical layering of the atmosphere implying that westerly transportation processes are not as stable and persistent as during advection from an easterly direction.

To sum up, the comparison of the approaches to trace methane sources/relative sinks gives results similar to the analogous comparison for CO_2. Deficits of the CWT analyses with areas remote from the considered receptor observatories are somewhat more apparent for CH_4. In the narrower catchment area as well as in the basic structures the very good correspondence of both models is confirmed. Taking into account the different methodical approach of the CO_2 and CH_4 budget estimations for the detection of relevant source and sink regions on the basis of measurement data versus inverse modelling for the quantification of the global climate gas fluxes, the very clear similarities of both models suggest that our methodology is well reliable for our purposes.

Conclusions and outlook

The synopsis of the previous chapters implies a high level of functionality and reliability of the modelling methodology we use to characterise the Alpine CO_2 and CH_4 budgets on the basis of atmospheric measurement time series from the observatories situated there. Regarding the clear parallels that can be seen in comparison with results from the inverse modelling of climate gas fluxes and concentrations—such as those derived from the Copernicus Atmosphere Monitoring Service (CAMS) and the Jena CarboScope project—the methodology of our study can be considered well capable of reliably detecting climate gas-specific source and relative sink regions with influence on the measurements at the Alpine receptor sites. The reliable project results, in conjunction with the positive results of the model's internal uncertainty estimates and external plausibility checks, highlight the model accuracy of the approach and, above all, underline the model's strength in accurately mapping spatiotemporal variations of the relevant emitters and absorbers of different climate gases (CO_2 and CH_4).

Thus, the highest positive contributions to the Alpine CO_2 concentrations derive from Eastern Europe, where the biggest European brown coal-mining areas are situated. With the same trustworthiness, we have identified typical CH_4 source and relative sink regions with southwestern Europe known by negative CH_4 contributions and emission-intensive regions in the north(west), the east and the south of Europe. The only exception to this northeast-southwest distinction is the Alpine region, which sticks out as a relative CH_4 sink from the surrounding source regions. Therefore, the Alpine core region represents a significant relative sink region for both examined climate gasses (CO_2 and CH_4) in the midst of partly emission-intensive Europe with regard to the study period and thus requires a special protection status also from the climate protection perspective.

An absolute prerequisite for a successful implementation of our model for the identification of potential emission hotspots and relative sink regions, together with their temporal variability, has been a sufficiently intensive coverage of the study region by the centroid pathways of the particle dispersion calculations. Only if the frequentation through the backward trajectories is high enough and the investigation period or the number of stations involved, respectively, is large enough for a sufficiently covered catchment area, the CWT analyses can produce meaningful maps. This restriction limits the application of the methodology to problems with low temporal resolution, which, for example, refer to seasonal or annual analyses, as in the present study. For the detection of relatively stationary source and sink regions and the variability of their influence over the course of the year, the method presented here on the basis of atmospheric measurement time series is very well suited, provided that an adequate data basis is guaranteed in the form of the long-term measurement series of a station or shorter measurement series of several stations covering at least a few years.

Taking into account, these requirements and the associated limitations of the methodological approach, the method presented fulfils the objectives of the study, as the plausible results of the fourth chapter and their comparison with largely comparable models demonstrate. The project results in form of the CWT maps attest the methodology its usefulness in answering the study's scientific questions, bringing out its strong advantages such as the high spatial resolution (0.2×0.2 degrees) or the climate gas specificity. Another benefit of the model is that it does not require a priori emission data, which means that the resulting outputs can possibly be used as an additional option for independent top-down plausibility checks and hence verification of bottom-up emission inventories. In order to be able to review the emission statistics, most of which are produced nationally, using the project's methodology, it is necessary to transform the database into a study area within national borders, in compliance with the requirements outlined above. The results presented here for the transnational study region of the Alps suggest a potentially promising transfer of the project approach to areas within national borders for the purpose of an independent validation of a country's emission inventories and open up future application possibilities for the model following the presented ones.

Abbreviations

GAW: Global Atmosphere Watch of WMO/UNO; GCOS: Global Climate Observing System of WMO/UNO; CAMS: Copernicus Atmosphere Monitoring Service NOAA Earth System Research Laboratory archive (NOAA CCGG); WDCGG: World Data Centre for Greenhouse Gases; RAMCES: Reseau Atmospherique de Mesure des Composes a Effet de Serre database; ICOS-ATC: Intergrated Carbon Observation System—Atmospheric Thematic Centre; NOAA: National Oceanic and Atmospheric Administration (USA).

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Authors' contributions

EG designed the computational framework of the study, derived the model and analysed the data. LR developed the basic idea for the project and provided all the technical details concerning the measurements of the CO_2 and CH_4 concentrations in compliance with the strict GAW standards. SH performed the particle dispersion runs with FLEXPART at the Leibniz Supercomputing Centre (LRZ) and was the expert for all matters related to the LPDM simulations. JJ supervised the project and contributed to the interpretation of the results. EG designed the figures and wrote the manuscript with input from all authors. The whole team of authors provided critical feedback and helped shape the research and analysis. All authors read and approved the final manuscript.

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Availability of data and materials

Data and material are available from the GAW WDCGG and the German Environment Agency.

Ethics approval and consent to participate

Not applicable.

Consent for publication

It is declared that a consent for publication exists.

Competing interests

The authors declare that they have no competing interests.

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