



**An airborne
perfluorocarbon
tracer system and its
first application**

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An airborne perfluorocarbon tracer system and its first application for a Lagrangian experiment

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Abstract

A perfluorocarbon tracer system (PERTRAS), specifically designed for Lagrangian aircraft experiments, has been developed by the Deutsches Zentrum für Luft- und Raumfahrt (German Aerospace Center, DLR). It consists of three main parts: a tracer release unit (RU), an adsorption tube sampler (ATS) and a tracer analytical system. The RU was designed for airborne tracer release experiments; meanwhile, it can be used on various platforms for different experimental purpose (here research vessel). PERTRAS was for the first time applied in the field campaign Stratospheric ozone: halogen impacts in a Varying Atmosphere (SHIVA) in November 2011. An amount of 8.8 kg perfluoromethylcyclopentane (PMCP) was released aboard the research vessel Sonne (RV Sonne) near the operational site of this campaign, Miri, Malaysia, on 21 November. The tracer samples collected using the ATS on board the DLR research aircraft Falcon were analyzed in the laboratory using a thermal desorber/gas chromatography/mass spectrometry (TD/GC/MS) system. Guided by forecasts calculated with the Lagrangian model, Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT), 64 tracer samples were collected onboard the Falcon approximately 5 and 25 h after the release, respectively, mostly with a time resolution of 1 min. Enhanced PMCP concentrations relative to ambient PMCP background values (mean: $6.62 \text{ fmol mol}^{-1}$) were detected during three intersects of the fresh tracer plume (age 5 h), with a maximum value of $301.33 \text{ fmol mol}^{-1}$. This indicates that the fresh tracer plume was successfully intercepted at the forecasted position. During the second flight, 25 h after the release, the center of tracer plume was not detected by the sampling system due to a faster advection of the plume than forecasted. The newly developed PERTRAS system has been successfully deployed for the first time. The instrumental set-up and comparisons between the measurements and HYSPLIT simulations are presented in this study.

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1 Introduction

Lagrangian experiments are a powerful tool in atmospheric investigations of the evolution of aerosols, clouds, and reactive chemical species during transport (Huebert et al., 1999). The approach in Lagrangian experiments is to measure the change of the composition of a single air mass as it moves using successive aircraft flights. A number of successful airborne Lagrangian experiments in the free troposphere have been performed using ambient tracer including ASTEX/MAGE (Clarke et al., 1996), ACE-1 (Bates et al., 1998), COBRA (Lin et al., 2004), and ICARTT (Fehsenfeld et al., 2006; Methven et al., 2006). However, it was recognized during these experiments that the ability to quantify the microphysical and chemical processes during transport could be greatly enhanced with the use of an artificial tracer for non-ambiguous air mass identification and information on mixing processes.

Perfluoroalkylcycloalkanes (hereafter PFCs) are regarded as ideal tracer compounds for the investigation of atmospheric transport and dispersion because they are non-toxic, non-reactive, non-scavenging, and non-depositing, and contribute neither to stratospheric ozone depletion nor to tropospheric air pollution. Besides, PFCs have also been used for many other applications including detection of fluid leaks in underground pipes, testing of effectiveness of underground storage tanks and covered waste sites, and determination of indoor air exchange rates. Furthermore, PFCs have a limited industrial use and thus have a low atmospheric background concentration, which is important for long-range Lagrangian experiments. The observed background concentrations of most PFCs used are at mixing ratios of typically a few parts per quadrillion (fmol mol^{-1}) (Piringer et al., 1997; Straume et al., 1998; Kim et al., 2002; Simmonds et al., 2002; Watson et al., 2007). They are potent greenhouse gases with a high global warming potential (GWP of about 7000). However, due to the very small atmospheric burden their global warming impact is insignificant compared to other greenhouse gases (Watson et al., 2007).

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PFCs can be detected down to background concentrations by enrichment on a solid adsorbent, separation by gas chromatography (GC), and detection using electron capture detection (ECD) (DeBortoli and Pecchio, 1985; Dietz, 1986; Lagomarsino, 1996; Straume et al., 1998). In recent years, due to the excellent selectivity and sensitivity and simple manipulation process, the analytical method with negative ion chemical ionization mass spectrometry (NICI-MS) for PFCs has increasingly been used (Begley et al., 1988; Galdiga and Greibrokk, 2000; Cooke et al., 2001; Simmonds et al., 2002).

The major PFC tracers, such as PMCP, perfluoromethylcyclohexane (PMCH), and perfluorodimethylcyclohexanes (PDCHs), are liquids with the appreciable vapor pressure and relatively low boiling point at atmospheric temperature and pressure. They can be released into the atmosphere by either atomization or vaporization followed by dilution to prevent condensation until the vapors are sufficiently diluted below the ambient dew point concentration (Dietz, 1986). In previous experiments, the PFCs were released by being directly introduced into the fast-moving air stream (Ferber et al., 1981) or through the evaporation after increasing the temperature above the tracers' boiling point with a heater (Draxler, 1985; Draxler et al., 1991). Afterwards, adsorbent samplers, as the most popular established technique for sampling PFCs were used to collect the tracer gas in the air (Ferber et al., 1981; Dietz, 1986; Draxler et al., 1991). So far mainly ground-based tracer experiments using PFCs have been performed in North America and Europe at continental and regional scales (Ferber et al., 1986; Draxler and Heffter, 1989; Ambrosetti et al., 1998; Nodop et al., 1998; Green, 1999; Johnson et al., 2000; Green et al., 2003). During these experiments the PFCs have been released from ground sites and ships and subsequently measured at different ground stations. Although airborne measurements (Ferber et al., 1986; Ambrosetti et al., 1998; Nodop et al., 1998; Johnson et al., 2000; Green et al., 2003) were also conducted for studies of the vertical mixing and dispersion of PFC-tagged air mass in some campaigns, these experiments were focused on the tracer transport near the ground. So far, no PFC tracer release and sampling system suitable for a development onboard

research aircraft in the free troposphere (FT) and the upper troposphere/lower stratosphere (UT/LS) have been reported.

In this paper, we present a new perfluorocarbon tracer system, PERTRAS, which was developed especially for airborne tracer release and detection. Two newly constructed automatic modules are parts of the system: a PFC release unit (RU) and an adsorption tube sampler (ATS). Combined with the well-established analytical technique of TD/GC/NICI-MS, the PERTRAS system was applied for the first time in the field campaign SHIVA. Section 2 gives a detailed description of the system. The tracer experiment during SHIVA is described in Sect. 3. In Sect. 4, the results of the first of the application of PERTRAS are presented. Finally, a summary and conclusion is given in Sect. 5.

2 Instrumental setup

A very first prototype of an airborne PFC release and sampling system was built in cooperation with the Brookhaven National Laboratory, and used for a pilot tracer experiment. However, leakage problems and aircraft certification issue required the construction of new RU and ATS modules, specifically designed for airborne operation. Here, stainless steel adsorption tubes are used for the sampler which are completely compatible with most commercially available Thermal Desorber system. In the next subsections, the instrumental setup of the PFC RU and ATS and the post analysis process in the laboratory are described in more detail.

2.1 PFC release unit (RU)

The basic principle of the PFC RU is to release the tracer liquid by spraying aerosol into a gas flow in the atmosphere through the process of high pressure atomization using a spray nozzle. Figure 1 shows a schematic diagram of the design of the PFC RU (left) and the aircraft module (right). It consists of three 6.6 L (~ 10 kg liquid PFCs

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each) stainless steel tanks. Depending on the planned experiments, up to 30 kg in total and three different PFCs can be released. The release rate can be set between 0.1 and 500 mL min⁻¹ with an uncertainty of 2 % using a High Pressure Liquid Chromatography (HPLC) pump (Knauer, Germany, type 1800). The pressure in the tube to the PFC outlet is adjustable between 40 and 100 bar. A 500 mL stainless steel cylinder of nitrogen (grade 5.0, Praxair) is installed ahead of the PFC tanks, and during release, nitrogen is used for equalization of the pressure in the tanks. The operation of the RU is achieved by using an onboard PC running LabVIEW through a touch screen control panel, or by remote controlling from a laptop through a Virtual Network Computing (VNC). The different PFC tanks can be selected by switching a three-way valve through controlling the solenoid valves (Fig. 1, left, solenoid valve 1). Then the release can start automatically by pressing the button on the controlling panel after a release program is set up. Three spray nozzles are used to disperse PFC droplets with diameters less than 20 µm into the fast-moving airstream. They are selected by controlling the solenoid valves (Fig. 1, left, solenoid valve 2), which are connected to the outlet tube of the HPLC pump. The HPLC pump maintains the pressure necessary for the nozzles to operate in the range where the droplets will be within this size range needed to avoid sedimentation of the droplets before evaporation. The RU has a total weight of 47 kg.

During the release operation, the small PFC droplets are dispersed into the wake of the aircraft or ship or the air flow at the ground. When the release is performed aboard an aircraft, the RU is installed in a rack in the aircraft cabin and the PFC release is through a special outlet mounted into the fuselage. For security reason, an additional container is integrated in the rack capable of housing the total amount of PFCs in case of a leak during flight. The PFC RU can both be used for ground-based and ship-based releases as well, depending on the different objectives of the experiments. During SHIVA the latter release platform was chosen, and the PFC RU (65.0 cm × 48.5 cm × 35.5 cm), shown in Fig. 1 (right), was installed on the deck of the stern of the research vessel (RV) *Sonne*. The sprayer was fixed at the pillar of the gunwale about 10 m above the sea surface.

2.2 Aircraft adsorption tube sampler (ATS)

The components of the aircraft ATS are shown schematically in Fig. 2. This sampler was constructed specifically for the purpose of airborne sampling. It consists of four 16-position multiport valves (Valco Instruments Co. Inc.). The sampler can collect maximum 64 samples with the standard adsorption tubes (outer diameter: 6.35 mm, inner diameter: 5 mm, length: 89 mm, material: stainless steel) continuously or selectively according to the experimental requirements. The four multiport valves are incorporated into a modified metal bellow pump (Ansyco, Germany). The flow rate is controlled by a metal mass flow controller (MFC, Bronkhorst, USA). The selection of the sampling tube is operated by the touch screen of an integrated aircraft ATS controller. Depending on different experimental purposes, the sampling flow, sampling time and the interval between two samples, etc., can be controlled through the touch screen as well. After a sampling sequence is programmed, the system runs automatically.

The gas flow has two different modes: standby/waiting and sampling. When the sampler is in the mode of standby (ready for sampling) or waiting for sampling (during the time intervals between two samples), ambient air is drawn into the system by the metal bellow pump (Fig. 2, through the dashed lines) directly without flowing through the adsorption tubes. When the ATS starts to sample, ambient air is drawn into selected adsorption tubes and the sampling flow is controlled by a MFC. Different amounts of sample air can pass through the tubes at optional intervals, and perfluorocarbon tracer molecules are trapped and concentrated in the tubes onto adsorbent material. The volume and the time of air passed through the tubes are recorded. After one sampling sequence is finished, the ATS is switched back to standby mode automatically. Before installing the ATS into an aircraft, the test of airtightness for each tube is conducted at the ground with high purity PFC-free nitrogen. During the field experiment SHIVA, the ATS (53.0 cm × 48.5 cm × 16.0 cm, 23.6 kg) (Fig. 3) was installed in the DLR research aircraft Falcon. The inner part of the ATS module is depicted in Fig. 3 (left). Figure 3 (right) shows the ATS integrated in the aircraft rack together with ozone and CO instruments.

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Stainless steel adsorption tubes (PerkinElmer, USA) are used to collect PFCs, filled with about 200 mg Carboxen 569 (20–45 mesh, Supelco, USA). After filling with Carboxen 569, all sampling tubes are conditioned at 260 °C for at least 5 h under a constant flow of high purity Helium. The tubes are periodically checked by GC-MS to ensure that they are free of PFCs before sampling.

Using adsorption tubes for the collection of gas samples, the suitable sampling volume related to the breakthrough of the analytes on the adsorbent material needs to be considered. There are many different definitions of breakthrough volume (BTV). It is usually defined as the gas volume which passes through the adsorbent bed before a given compound begins to be eluted from the sorbent. Different sample volumes were tested to ensure that no PFC tracer had broken through the adsorbent bed during sampling. For this, the first blank adsorption tube of Carboxen 569 was loaded with a certain amount of gas PFCs standard. This tube was connected in series with another blank tube to retain the PFCs eluted from the front to the back sorbent tube (Aragón et al., 2013). The results showed that there was no PFC penetration with 40 L PFC-free nitrogen at the flow rate of 200 mL min⁻¹ flushing through the adsorption tubes at 20 °C. The theoretical BTV of these sample tubes with Carboxen 569 should be higher than 40 L for most PFCs. In addition, according to the results of Yang et al. (2005), the penetration volume of perfluorodimethylcyclobutane (PDCB) is almost independent of the flow rate (≤ 1.5 L min⁻¹) of the mobile phase. Other PFCs have stronger adsorption ability than PDCB on Carboxen 569. Therefore, several liters of PFCs with less than 1.5 L min⁻¹ flow rate can be trapped onto the sampling tube without exceeding the theoretical BTV.

2.3 Analytical instruments

The identification of the PFCs and quantification of their concentrations is accomplished offline by using an analytical system consisting of a GC (Tracer GC Ultra) connected with a pre-concentrated thermal desorber (TD) and a mass spectrometry (MS) in the laboratory. Here only a brief description of the analytical process is given. More

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details are given by Ren et al. (2014). Thermal desorption was carried out with a thermal desorber ATD650 (PerkinElmer, USA). The ATD650 was connected to the injection port of the GC through a temperature controlled interface. With a two-step desorption, the sample which has been enriched into the cold trap, maintained at a temperature of -30°C , can be desorbed in $\sim 3\text{ s}$ onto the analytical column in GC (Tracer GC Ultra) oven in split mode. The cold trap was filled about Carboxen 569 (20–45 mesh, Supelco, USA) and conditioned by heating at 260°C with carrier gas for at least 5 h under high purity Helium to clean prior to the normal analysis. The temperature of both the interface and the valve was set at 200°C .

The separation of the tracer was carried out on a $30\text{ m} \times 0.25\text{ mm} \times 5.0\text{ }\mu\text{m}$ Al_2O_3 -PLOT-S capillary column (HP-PLOT Al_2O_3 S, Agilent, USA). A particle trap $5\text{ m} \times 0.25\text{ mm}$ deactivated Fused Silica column (Agilent, USA) was connected to the Al_2O_3 -PLOT-S to avoid particles shedding from the stationary phase layer of PLOT capillary column entering the MS detector. The following oven temperature program was set up: 100°C (0.5 min), $30^{\circ}\text{C min}^{-1}$ to 130°C (6.5 min), $30^{\circ}\text{C min}^{-1}$ to 140°C (23 min), then $20^{\circ}\text{C min}^{-1}$ to 190°C (5 min). High-quality helium (AlphagazTM He, 6.0, Air Liquide) was used as carrier gas with a flow rate of 1 mL min^{-1} . The mass spectrometry was operated in negative ion chemical ionization (NICI) mode at 70 eV and in selected ion monitoring (SIM) mode. Methane (N55 quality, Air Liquide) was used as reagent gas. The GC/MS interface and the source temperature were set to 190°C . It has been demonstrated by Ren et al. (2014) that this detection system can be used for routine laboratory analysis of several PFCs with good sensitivity and selectivity including PMCP, PMCH, PDCH, PDCB and PECH (perfluoroethylcyclohexane).

Samples were calibrated using a gas standard prepared at the University of Bristol (Simmonds et al., 2002). The limit of detection (LOD) of the method for PMCP was 1.2 ppq (fmol mol^{-1}), detected with a 3 times signal-to-noise ratio. The precision of PMCP during the analytical process was $\pm 4.6\%$. The calibration appeared to be linear at least 3 orders of magnitude for the concentrations that were studied.

3 Field Deployment of PERTRAS

3.1 SHIVA – measurement campaign

SHIVA is a European Union (EU) funded research project (<http://shiva.lup.uni-heidelberg.de/>) with the main objective to reduce uncertainties in present and future stratospheric halogen loading and ozone depletion. Of particular relevance are studies of short and very short-lived substances (VSLS) with climate-sensitive natural emissions. A core field campaign was conducted during November–December 2011 in the tropical Western Pacific region with measurements along the coastline of the Peninsula Malaysia and Borneo using ships and aircraft. The campaign base of the DLR research aircraft Falcon was Miri on Borneo, Malaysia. One measurement objective was to study the transport and dispersion of VSLS halocarbons produced from seaweed along the coast of Borneo in the marine boundary layer and the entrainment of these emissions in the FT (Aschmann et al., 2009; Tegtmeier et al., 2012, 2013). SHIVA provided the opportunity to deploy the PERTRAS system for the first time in the framework of a Lagrangian experiment. In the following, the experimental details of the tracer release and collection are given.

3.2 Tracer release

The PFC tracer release was conducted from the German RV *Sonne*. Therefore the release unit with PMCP was set up on the deck of the *Sonne*. The time of the tracer release was selected based on HYSPLIT forecasts. An amount of 8.8 kg pure PMCP was released on 21 November 2011 from 02:24 UTC (112.96° E, 4.51° N) to 03:03 UTC (113.01° E, 4.56° N). The release position along the *Sonne* route is depicted in Fig. 4 as site C4.

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3.3 HYSPLIT simulation and the flight patterns

Version 4 of HYSPLIT (Draxler and Hess, 1997, 1998; <http://www.arl.noaa.gov/ready/hysplit4.html>) was employed to forecast and simulate the transport and dispersion of the PFC tracer and to support flight planning. Data from the Global Forecast System (GFS, for forecast) and Global Data Assimilation System (GDAS, for analysis) with $1^\circ \times 1^\circ$ horizontal resolution, and 3 h time intervals were used as meteorological input for HYSPLIT. Data were given on pressure-levels with a resolution of 25 hPa within the lower atmosphere. HYSPLIT was operated in particle dispersion mode and all parameterizations were set to the standard values. The release species was treated as a passive gas tracer without deposition. The simulation takes account of the movement of the ship during the release, that is, the release is split into 39 point-sources (one for each minute) shifted in space and time according to the ship course. The HYSPLIT forecast was regularly updated every 6 h based on the latest available GFS data.

Figure 5 shows HYSPLIT forecasts of tracer concentrations after PMCP was released from the RV Sonne on 21 November 2011 over a dispersion period of 5 and 25 h, respectively. The forecasted maximum concentration in the PMCP tracer plume is depicted in dark red. Based on these forecasts, the corresponding flight plan of the Falcon at these two days was made and the flight tracks for the two flights are also illustrated in Fig. 5.

3.4 Tracer sampling

The ATS was successfully deployed on board the Falcon during two flights conducted on 21 and 22 November 2011, respectively. The aim was to test and verify the operation of the whole analytical system and to compare the tracer measurements with the HYSPLIT simulation. Therefore, multiple Falcon transects were planned through the center of the tracer plume based on the HYSPLIT forecasts, as shown in Fig. 5. In general, the collection of samples was started several minutes before the expected entering of the tracer plume according to the forecast, and sampling was terminated a few

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minutes after the expected exit of the plume. On 21 November, the Falcon performed two intersection of the plume rotated by 90° , at two altitudes (170 m and 500 m), about 75 km away from the release point. Before flying back to Miri, the aircraft ascended to about 1000 m, to try to catch the highest plume part according to the vertical cross-section map. On 22 November, the Falcon flew through the elongated plume at about 800 m and 1200 m, respectively, about 150 km away from the release point. All samples were collected at 1 L min^{-1} for 1–1.5 min. There was 0.5 to 2 min intervals between two samples in a sampling sequence, and the intervals were adjusted due to the actual sampling situation. During the sampling periods, the aircraft speed ranged between $101\text{--}112 \text{ m s}^{-1}$ (21 November) and $123\text{--}140 \text{ m s}^{-1}$ (22 November), thus 1–1.5 min collection times result in a horizontal grid spacing of about 6–10 km and 7–13 km, respectively. In accordance with the flight plan, totally 64 samples were obtained in these two flights, including 4 blanks for quality control to check if any contamination of the sampling tubes had taken place during transport, sampling or analysis. To avoid contamination, the sampling tubes had been separated strictly from the tracer storage tank and the tracer releaser system during the whole campaign.

In addition, a commercial sequential tube sampler (STS 25, PerkinElmer, USA) was used to collect background samples at a rural site (112.978° E , 4.398° N) near the airport of Miri, about 9 km south of the center of Miri. Using a sampling flow rate of 200 mL min^{-1} , air samples were trapped onto adsorption tubes for 40 min. A total of 37 samples were collected between 20 November and 5 December 2011, including 8 blanks, which were also used for quality control purpose. Since the plume of the released PMCP tracer was not advected to Miri, background samples could still be collected at the ground in Miri after the release experiment. Before and after the ground-based sampling, the flow through every sampling orifice of the ground sampler was measured with a Gillian Galibrator-2 (Scientific Instrument Services, IncTM, USA). After the sampling period, the tubes were sealed immediately at both ends with 1/4 inch brass storage caps with 1/4 inch PTFE ferrules and stored until analysis.

4 Results and discussion

The analysis of all blank tubes showed that no contamination occurred during the transport of the tubes to and from the campaign site, and during sampling, storage and analysis. The measured background concentrations of PMCP at the ground near Miri range between 5.18 and 8.95 fmol mol⁻¹, with a mean value of 6.62 fmol mol⁻¹ and a standard deviation of 1.02 fmol mol⁻¹ (6.62 ± 1.02 fmol mol⁻¹). This is comparable to PMCP levels reported from previous field experiments (Kim et al., 2002; Simmonds et al., 2002; Watson et al., 2007; Ren et al., 2014). In the following subsections, the analytical results of the tracer measurements with PERTRAS are presented and comparisons between the measurement and HYSPLIT simulation are discussed.

4.1 Time series of PMCP measurements

Figure 6a shows a time series of PMCP tracer concentrations (red bars) along with altitude (in black) for the flight on 21 November 2011. In total, 33 air samples were collected during five cross-sections at three different flight levels (170, 500 and 1000 m). The measured PMCP concentrations range from 9.36 to 301.33 fmol mol⁻¹, largely exceeding the mean PMCP background concentration measured at the ground (6.62 fmol mol⁻¹) for most samples. Three time periods of clearly elevated PMCP concentrations are observed for the first three plume crossings during 07:00–07:25, 07:45–08:05, and 08:25–08:45 UTC (about 5 h after the release), at altitudes of 170 m, 170 m and 500 m, respectively. This demonstrates that the Falcon successfully sampled the tracer plume during these periods.

During the first sampling period at 170 m from northeast (NE) to southwest (SW), three samples collected between 07:06 and 07:15 UTC showed enhanced tracer concentrations relative to background concentrations. The maximum detected PMCP concentration (301.33 fmol mol⁻¹) at 07:07 UTC is about 45 times higher than the mean background at the ground. Along both sides of the maximum, the detected PMCP concentrations decrease reaching values close to the background level. At around

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07:10 UTC there is a 5 min sampling gap, due to a short period of malfunction of the sampling pump. During this time interval, two samples could not be taken. During the second sampling period (07:45–08:05 UTC), when the Falcon flew from northwest (NW) to southeast (SE), most samples were clearly higher than the background with a maximum value of $203.39 \text{ fmol mol}^{-1}$. The expansion of the tracer plume in NW–SE direction is found to be broader than along the perpendicular direction (NE–SW), and the concentration variation of PMCP are comparatively moderate. During the third sampling period at around 08:25–08:45 UTC, when the Falcon flew along the NE–SW direction again but at 500 m, the PMCP concentrations show a quasi-symmetrical distribution. Very likely the Falcon flew through a narrow region of the tracer plume or just touched the fringe of the tracer plume. During the last sampling period from 09:05 to 09:25 UTC two of the samples have slightly higher concentrations than the background. The samples collected at 1000 m (09:45–10:00 UTC) have PMCP concentrations close to background values. Probably the Falcon flew above the tracer plume here.

For the tracer sampling flight on 22 November, about 25 h after the release, a total of 27 samples, all except one, were successfully analyzed. The corresponding time series of PMCP concentrations is shown in Fig. 6b (purple bars). Most PMCP values detected (maximum concentration of $20.14 \text{ fmol mol}^{-1}$) are higher than background. Unfortunately, the tracer plume was advected more south than forecasted and partly over land. Therefore the lowest level of the Falcon possible was 1000 m and the sampling occurred only across the outer edge of the tracer plume.

4.2 Comparison between the measurement and HYSPLIT simulations

The comparisons between the PMCP measurements and the HYSPLIT simulations based on GDAS data for the flight on 21 November show in general a reasonable good agreement as illustrated in Fig. 7.

During the first sampling period (marked as “1” in Fig. 7) at around 07:10 UTC, the peak PMCP concentration simulated by HYSPLIT ($6190.74 \text{ fmol mol}^{-1}$) is much higher than the maximum PMCP concentrations observed ($301.33 \text{ fmol mol}^{-1}$)

(07:06–07:07 UTC), however, the HYSPLIT tracer maximum is calculated for the period where two samples could not be taken due to the short pump failure. The average PMCP concentration ($279.31 \text{ fmol mol}^{-1}$) calculated with HYSPLIT for the period 07:06–07:07 UTC is similar with the measurements.

During SHIVA, the University of Frankfurt operated also an in-situ gas chromatograph GHOST (Gas-chromatograph for the Observation of Stratospheric Tracers-coupled with a mass spectrometer) on board the Falcon for measurements of halogen compounds (Bujok et al., 2001; Sala et al., 2014). GHOST was not prepared or calibrated to measure PFCs, however detected signals of PMCP during the tracer plume sampling.

Interestingly, during the sampling period 07:00–07:15 UTC, three high signals of the PMCP tracer were detected by GHOST, one being extraordinarily high. One of the GHOST signals well correspond in time with one of the PERTRAS samples during the period 07:12–07:13 UTC (the third bluish-green bar, Fig. 7). Based on the ratios of both signals to the calibration gas used for GHOST, the PMCP concentrations of the two other GHOST signals can be estimated collected from 07:03 to 07:04 UTC, between the first and the second sample of ATS, and from 07:07 to 07:08 UTC, starting just after the sample from ATS being finished at around 07:07 UTC. The GHOST estimates for the PMCP concentrations amount to 46.8 and $840.6 \text{ fmol mol}^{-1}$, respectively, and are also shown in Fig. 7 as bluish-green bars (the first and the second one). Overall the PMCP measurements and the HYSPLIT simulations indicate that the Falcon was traversing the center of the tracer plume during the first sampling period.

For further comparisons of the PMCP measurements and HYSPLIT simulations, time-snapshots of the tracer plume for each 10 min time interval for the sampling flight on 21 and 22 November are shown in Figs. 8 and 9. Here, the horizontal model layers are depicted which match best with the Falcon flight altitude during each 10 min time window from $t - 5 \text{ min}$ to $t + 5 \text{ min}$ for the corresponding samples. For those cases where the flight altitude varied by more than 100 m during the corresponding time window, the contours show, at each horizontal grid point, the highest volume mixing ratio (VMR) of

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any vertical model layer which the aircraft crossed. Those height ranges are indicated in the headings of the plots.

As shown in cases a and b of Fig. 8, which are corresponding to the sampling period of “1” in Fig. 7, the flight track was almost passing through the center of the tracer plume as planned. In b, the samples collected at 07:07 (yellow oval) and 07:12 UTC (green oval), with PMCP concentrations of $301.33 \text{ fmol mol}^{-1}$ and $58.54 \text{ fmol mol}^{-1}$, respectively, are quantitatively similar to the simulation results. However, one sample (another green oval in case a) with a PMCP concentration of $62.58 \text{ fmol mol}^{-1}$, very well above background values, was collected out of the main forecasted plume. This indicates that the simulation is underestimating the size of the plume in NE–SW direction at least by 10 km. However, this difference can be regarded as reasonable since the model cannot reproduce such small variations in the measured tracer concentrations because it is below the resolution of the meteorological input data (Stohl et al., 1998).

For the other two sampling periods during 07:45–08:05 UTC (Fig. 7, case 2) and 08:25–08:45 UTC (Fig. 7, case 3), the simulated PMCP concentrations are similar in magnitude to the measurements. These samples were mainly collected in the boundary region of the plume. As can be seen from Fig. 8c, during 07:45–08:05 UTC, two samples, illustrated as light green and yellow ovals, with PMCP concentrations of $179.66 \text{ fmol mol}^{-1}$ and $203.39 \text{ fmol mol}^{-1}$, respectively, show reasonable agreement with the model, but are located in the boundary of the plume in the northeastern direction. The plume was transported a little bit further to the south compared to the forecast used for flight planning. Two other samples illustrated as light green ($159.36 \text{ fmol mol}^{-1}$) and green ovals ($51.96 \text{ fmol mol}^{-1}$) (Fig. 8d), are located outside of the simulated plume, which indicates that the plume in this area probably dispersed more (at least 12 km) in the SE direction than simulated by the model. From the reanalysis for the time period 08:30–08:40 UTC (Fig. 8e and f), the flight track seems to have touched the southwestern edge of the plume as speculated in Sect. 4.1. This implies that the tracer plume in reality was advected further to the southeast. Although the Falcon mostly did

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not fly through the plume center as planned, the results from the measurements (Fig. 7) coincidence with the HYSPLIT simulations quite well.

The results of samples collected during the fourth and fifth sampling period (marked as “4” and “5” in Fig. 7) at around 09:15 and 09:55 UTC, are not captures by the simulations. This suggests that the tracer plume moved away already from the sampling area, which can be seen more clearly in the Fig. 8g and h.

For the sampling flight on 22 November, if the analysis of HYSPLIT is correct, the Falcon sampled north of the main plume, barely touching the northeastern edge from 04:10 to 04:30 UTC (shown in Fig. 9). According to the HYSPLIT forecast, which was used for the flight planning, the tracer plume should have been located further north. Another difficulty for the flight plan was that the plume was located partly over land and requiring a minimum flight altitude of the Falcon possible was 1000 m. The latest updated HYSPLIT forecasts for the flight on 22 November after the take-off of the Falcon already showed that the tracer plume center was moving faster and more to the south, however, the flight track of the Falcon could not be changed due to telecommunication loss with the Falcon because of the low flight altitude. Therefore the Falcon collected samples according to the flight plan made before take-off. The results from the measurement basically confirm the post-flight simulation of HYSPLIT using analyzed meteorological data.

5 Summary and conclusion

We have presented a newly developed perfluorocarbon tracer system, PERTRAS, including a tracer release unit (RU), an adsorption tube sampler (ATS), and an analyzing system TD/GC/NICI-MS (thermal desorber/gas chromatography/negative ion chemical ionization-mass spectrometry). The PERTRAS modules are easily portable, can be automatically operated and are suitable for the deployment on different platforms (ground station, ship and aircraft) depending on the objective of the tracer experiment, although it was designed for the application aboard aircraft. The tracer release rate can

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be adjusted between 0.1 and 500 mL min⁻¹ with an uncertainty of 2 % using a HPLC pump and up to a total of 30 kg and three different PFCs can be released. Up to 64 tracer samples can be taken during a flight with the tracer sampler ATS. The sampling procedure can be programmed including sequences with standby/waiting and sampling modes. All sample tubes are analyzed with a high selective and sensitive TD/GC/NICI-MS system in the laboratory with a limit of detection of 1.2 fmol mol⁻¹.

During the SHIVA field campaign in November 2011, the new PERTRAS system was applied for the first time. The tracer release was conducted on the deck of RV *Sonne* and 8.8 kg PMCP was released during a period of 39 min (02:24 to 03:03 UTC). The tracer was sampled by the DLR research aircraft Falcon using the ATS during two flights on 21 and 22 November, approximately 5 and 25 h after the release, respectively. In total, 63 samples including 4 blanks were collected, with a time resolution of 1 min, and analyzed after the campaign in the DLR laboratory using a TD/GC/NICI-MS system. The result of all blank samples shows that no contamination occurred during the transport of the sampling tubes to the campaign site and back, as well as during sampling and analysis.

During the sampling flight on 21 November the tracer plume was intercepted successfully several times and a reasonable agreement was found between the tracer measurements and post-flight HYSPLIT simulations using analyzed meteorological fields. It could be demonstrated that the entire PERTRAS system worked reliable. The plume simulated by HYSPLIT is somewhat narrower than the one inferred from the measurement for the first three sampling periods for the 21 November flight implying a slight underestimation of dispersion and mixing by HYSPLIT.

The tracer plume was largely missed during the flight on 22 November due to several reasons. Firstly, there was a restriction for the minimum flight altitude of 1000 m since the tracer plume was advected partly over land. Secondly, a needed adjustment of the flight pattern after the Falcon take-off based on the latest HYSPLIT forecasts could not be transmitted to the Falcon crew due to a loss of telecommunication because of the

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low flight altitude. These problems should not occur for tracer experiments at higher altitudes.

After the successful deployment of PERTRAS during the SHIVA field campaign further tracer experiments are planned to study transport and transformation processes in deep convection in the tropics and in large scale pollution plumes of mega cities in Asia. Recently, the PERTRAS system has been applied successfully during the campaign T-NAWDEX-Falcon in a warm conveyor belt over Europe. Analysis of this data is on-going and results will be presented in a forthcoming paper.

Acknowledgements. This work was funded by the Deutsche Forschungsgemeinschaft (DFG) under Priority Program HALO SPP 1244 (SCHL 1857/1-2) and by the EU project SHIVA (EU-226224). We especially thank Michael Lichtenstern (DLR) for his excellent work of constructing PERTRAS instrumentation. We thank Russel N. Dietz and Thomas B. Watson of the Brookhaven National Laboratory for very helpful advice in all aspects of the PFC technology. We also thank Dominik Schäuble and Tina Jurkat (DLR) for the tracer release from the Sonne and Klaus Pfeilsticker and Marcel Dorf (University Heidelberg) for coordinating the SHIVA campaign. Stephan Sala and Andreas Engel (University Frankfurt) are acknowledged for providing the GHOST data. Thanks are also due to Damien Martin (University of Bristol) for providing the gas standard. We acknowledge the support of the pilots and staff of the DLR Flight Department and the help of Anke Roiger, Anja Reiter and Heinfried Aufmhoff (DLR) during the campaign. We are thankful to the NOAA Air Resources Laboratory for providing the HYSPLIT model.

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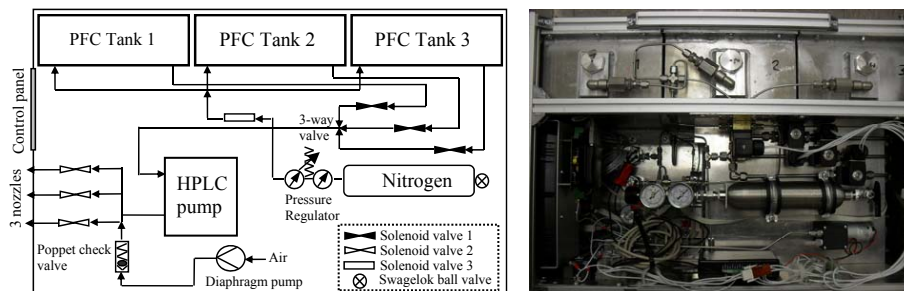


Figure 1. PFC Release Unit: schematic of gas flow (left); Aircraft release module (right).

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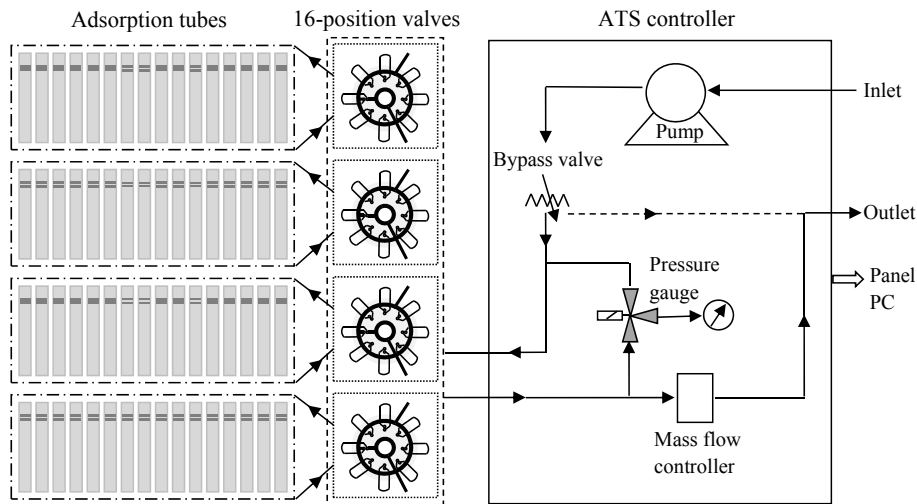


Figure 2. Schematic of the ATS. In standby or waiting mode, gas flow occurs through the dashed lines to the outlet. In sampling mode, gas flow is through the solid lines into the adsorption tubes.

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Figure 3. Adsorption tube set-up in the sampler ATS (left) and aircraft instrument rack including PERTRAS, ozone and CO instruments (right).

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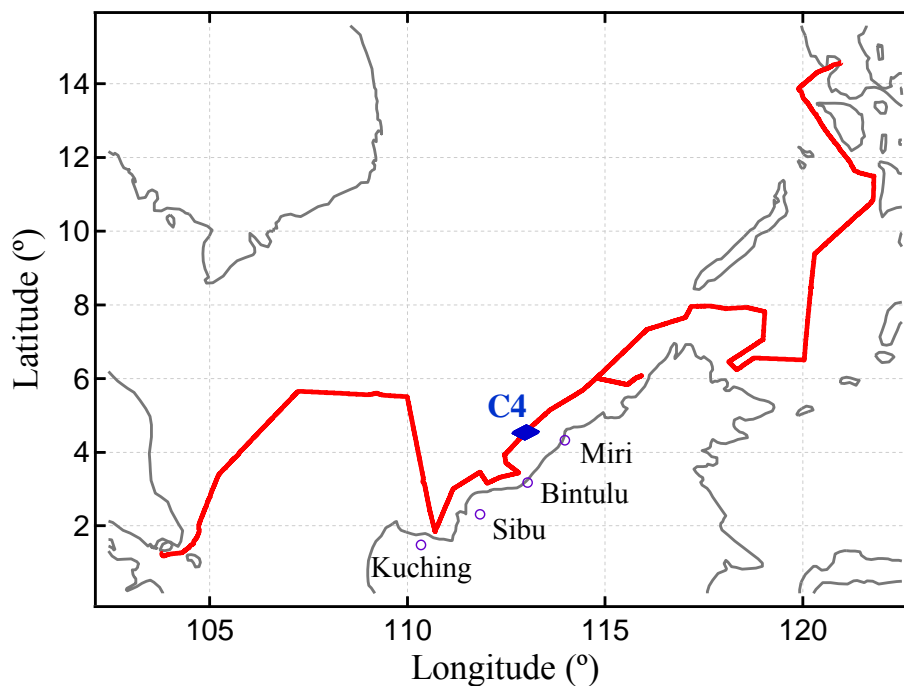


Figure 4. Route of the RV *Sonne* during SHIVA field campaign (red line) and the position (C4) of the tracer release: PMCP was released on the deck of *Sonne* from 02:24 to 03:03 UTC on 21 November 2011.

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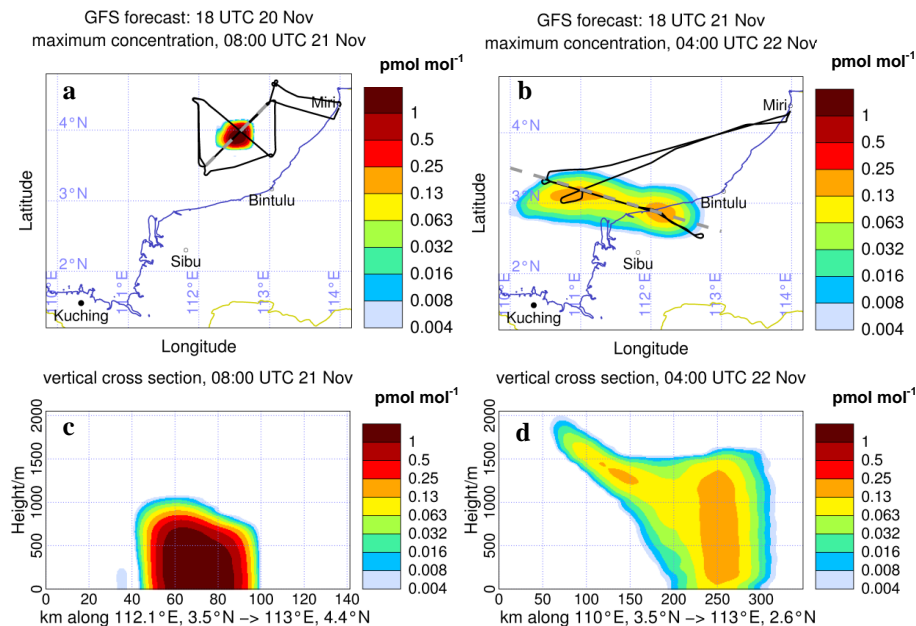


Figure 5. HYSPLIT forecasts of the horizontal and vertical distribution of the PFC concentration at the northern coast of Borneo, 5 h (a and c) and 25 h (b and d) after the release of the tracer. The flight track of the two Falcon sampling flights are also shown (black lines in a and b). The thick dashed lines (grey) indicate the location of vertical cross sections (c and d).

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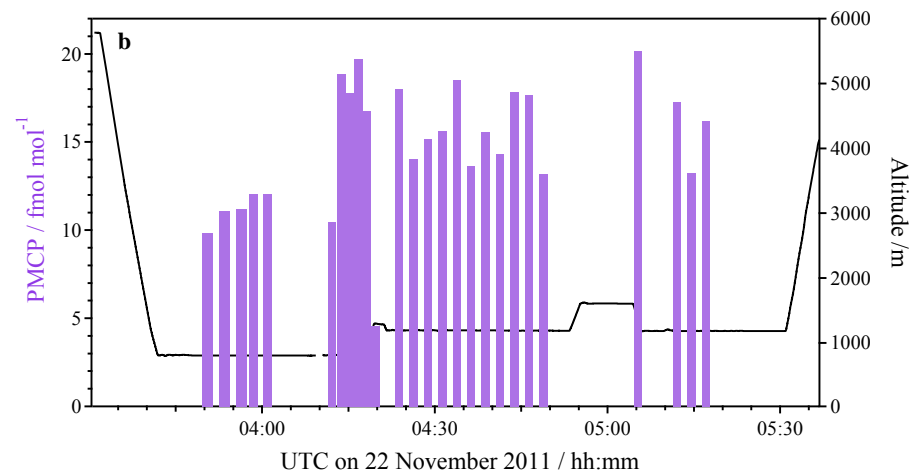
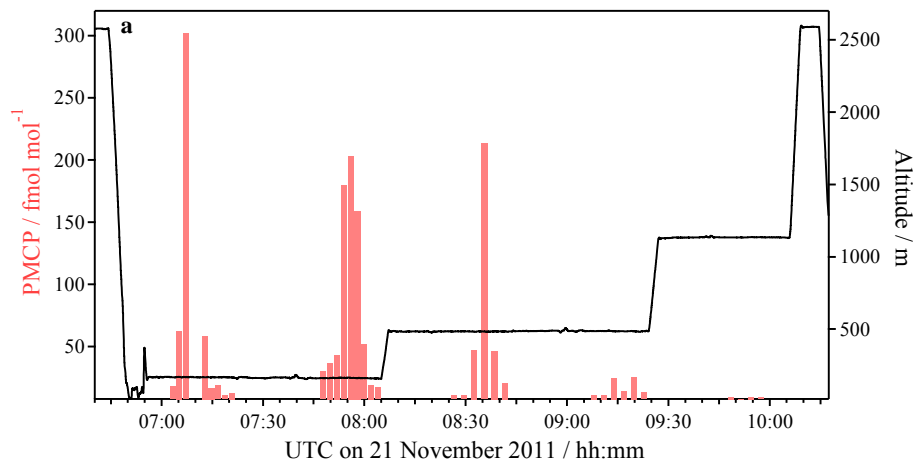


Figure 6. Time series of PMCP concentrations on 21 November (a) and 22 November 2011 (b). Each bar represents the averaged value during each sampling interval.

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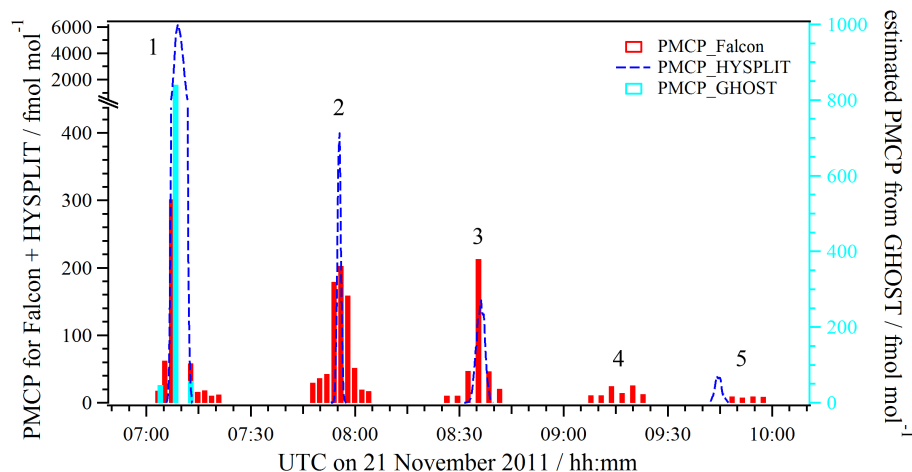


Figure 7. Time series of measured PMCP concentrations (red bars) on 21 November and HYSPLIT simulations using analyzed meteorological fields (blue dashed lines). Bluish-green bars represent the PMCP concentrations estimated from the signals of the GHOST instrument.

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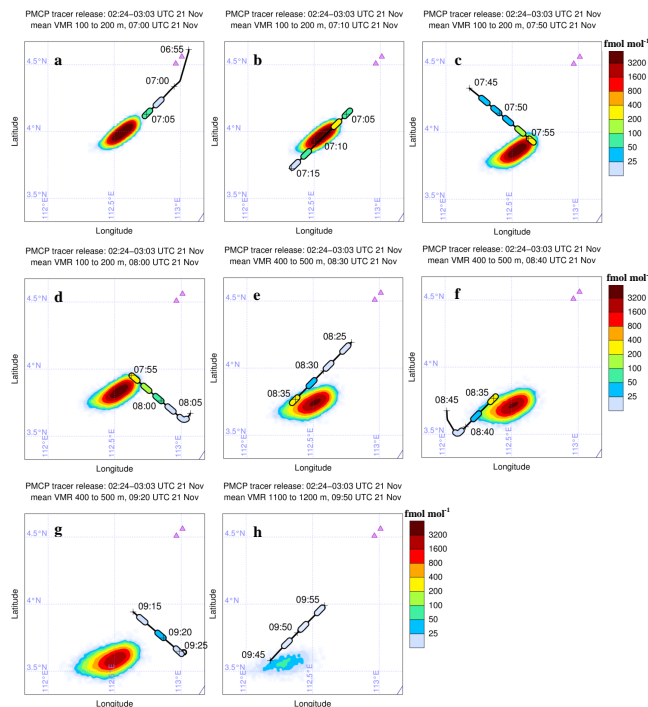


Figure 8. Time snapshots of the simulated tracer plume with HYSPLIT using analyzed meteorological fields with HYSPLIT using analyzed meteorological fields (big plume) on 21 November, and the corresponding sampling tracks (black line). (a and b) the plume at $t = 07:00$ and $07:10$ UTC, (c and d) the plume at $t = 07:50$ and $08:00$ UTC, (e and f) the plume at $t = 08:30$ and $08:40$ UTC, (g) the plume at $t = 09:20$ UTC, and (h) the plume at $t = 09:50$ UTC. The volume mixing ratio (VMR) is calculated as mean value within vertical layers of 100 m thickness, for (a–d) 100–200 m, for (e–g) 400–500 m, for (h) 1100–1200 m. Small colored ovals represent the samples collected along the flight track (same color scale as used for the HYSPLIT data). Two purple triangles indicate the positions of the start and the end of the tracer release.

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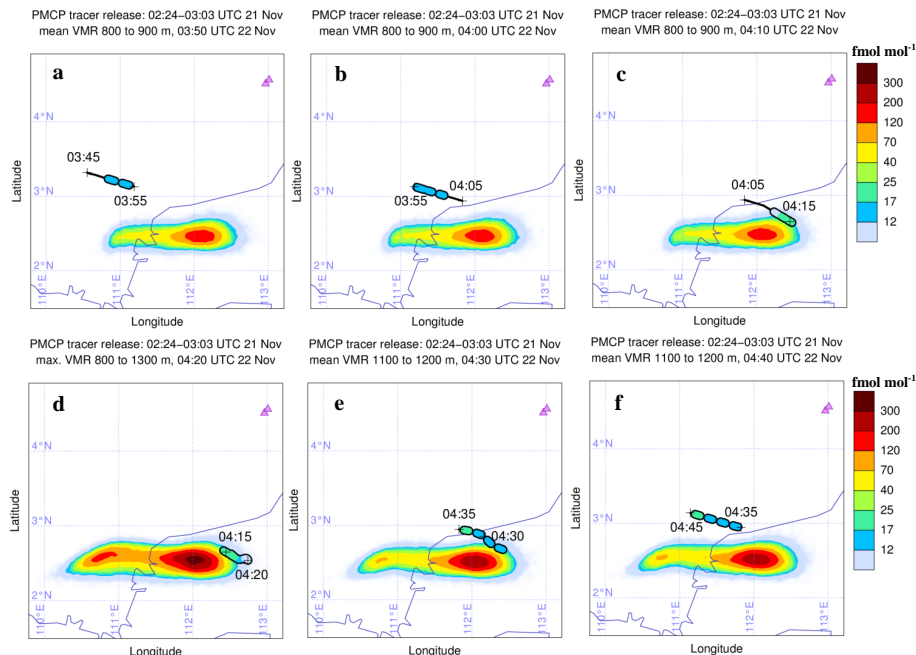


Figure 9. Time snapshots of the simulated tracer plume (big plume) on 22 November, and the corresponding sampling tracks (black line). **(a–f)** the plume at $t = 03:50, 04:00, 04:10, 04:20, 04:30,$ and $04:40$ UTC, respectively. VMR is calculated as mean value within vertical layers from 800–900 m **(a–c)** and 1100–1200 m **(e and f)**, respectively. **(d)** picks, at any location, the maximum out of 5 vertical layers between 800 m and 1300 m, because the flight altitude changed during this section. Small colored ovals represent the samples collected along the flight track (same color scale as used for the HYSPLIT data). Two purple triangles indicate the positions of the start and the end of the tracer release.

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