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Impact of stratospheric intrusions and intercontinental transport on ozone at Jungfraujoch in 2005: comparison and validation of two Lagrangian approaches

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Abstract

The particle dispersion model FLEXPART and the trajectory model LAGRANTO are Lagrangian models which are widely used to study synoptic-scale atmospheric air flows such as stratospheric intrusions (SI) and intercontinental transport (ICT). In this study, we focus on SI and ICT events particularly from the North American planetary boundary layer for the Jungfraujoch (JFJ) measurement site, Switzerland, in 2005. Two representative cases of SI and ICT are identified based on measurements recorded at Jungfraujoch and are compared with FLEXPART and LAGRANTO simulations, respectively. Both models well capture the events, showing good temporal agreement between models and measurements. In addition, we investigate the performance of FLEXPART and LAGRANTO on representing SI and ICT events over the entire year 2005 in a statistical way. We found that the air at JFJ is influenced by SI during 19% (FLEXPART) and 18% (LAGRANTO), and by ICT from the North American planetary boundary layer during 13% (FLEXPART) and 12% (LAGRANTO) of the entire year. Through intercomparsion with measurements, our findings suggest that both FLEXPART and LAGRANTO are well capable of representing SI and ICT events if they last for more than 12 h, whereas both have problems on representing short events. It is also shown that although the long-range transported air is characterized by relatively low NO_y/CO ratios and elevated CO concentrations, using a combination of NO_y/CO and CO as control parameters still encounters difficulty in distinguishing aged air masses by their source regions. Moreover, a sensitivity study indicates that the agreement between models and measurements depends significantly on the threshold values applied to the individual control parameters. Generally, the less strict the thresholds are, the better the agreement between models and measurements. Although the dependence of the agreement on the threshold values is appreciable, it nevertheless confirms the conclusion that both FLEXPART and LAGRANTO are well able to capture SI and ICT events with sustaining time longer than 12 h.

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1 Introduction

Tropospheric ozone (O_3) is known as the most important air pollutant of summer smog causing harm to human health and ecosystem (e.g., Brunnekreef and Holgate, 2002). It strongly determines the oxidizing capacity of the troposphere and acts as the third relevant greenhouse gas in terms of anthropogenic radiative forcing at the Earth's surface (Houghton et al., 2001). For these reasons, tropospheric O_3 concentrations have been an important subject of atmospheric research over the last decades. Tropospheric O_3 is known to have two sources: Chemical production by the oxidation of volatile organic carbons (VOCs) and carbon monoxide (CO) in the presence of nitrogen oxides (NO_x=NO+NO_2) (e.g., Chameides and Walker, 1973; Crutzen, 1974; Levy, 1971), and transport from the stratosphere (e.g., Danielsen, 1968; Junge, 1962). Many studies have tried to determine the contribution of O_3 from the stratosphere to tropospheric O_3 (Bourqui, 2006; Brioude et al., 2007; Cristofanelli et al., 2003; Hsu and Prather, 2005; James et al., 2003b). Even if it is currently thought that the greatest contribution to tropospheric O_3 comes from photochemical production (Staehelin et al., 1994; Yienger et al., 1999), the contribution from the stratosphere cannot be neglected (Roelofs and Lelieveld, 1997).

Recent studies have also suggested that cross-tropopause mass flux may increase during the 21st century as a result of climate change (Sudo et al., 2003; Collins et al., 2003). According to these studies, the long term enhancement of stratospheric O_3 could significantly modify the tropospheric O_3 budget. Ordóñez et al. (2005) suggested that the positive O_3 trends in the lower free troposphere over Europe during the 1990s were likely to a large extent due to enhanced flux of O_3 from the stratosphere. Tarasick et al. (2005) examined Canadian ozonesonde data and suggested that the correlation between O_3 changes in the lower stratosphere and in the troposphere at the Canadian ozonesonde stations is significant, leading to the conclusion that at least part of the changes in the troposphere results from changes in the lower stratosphere. All these findings indicate that stratospheric intrusions (SI) which transport O_3-rich air into the

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troposphere is one of the most important processes that impacts the tropospheric O_3 budget.

The lifetime of O_3 in the free troposphere ranges from a few days to several months (e.g., Liu et al., 1987), which allows its transport over distances of intercontinental and hemispheric scales. Transport of O_3 and O_3 precursors may thus have impacts on O_3 concentrations found downwind of industrialized regions of the Northern Hemisphere, where most of the O_3 precursors (VOCs and NO_x) are emitted (Berntsen et al., 1999; Jacob et al., 1999; Jonson et al., 2001; Stohl, 2001; Wild and Akimoto, 2001; Wild et al., 2004). In particular, transatlantic transport of O_3 is raising a lot of interests because of the relatively short distance between North America and Europe (Wild and Akimoto, 2001). For example, Li et al. (2002) reported that anthropogenic emissions from North America led to an additional 20% of violations of the European council ozone standard in summer 1997 over Europe. A recent study using the GEOS-CHEM global transport model also indicated that North American emissions contribute 11% to the tropospheric O_3 annual average total burden over Europe while European sources only contribute 9% (Auvray and Bey, 2005).

Trajectory models are widely applied to investigate atmospheric transport processes such as SI and intercontinental transport (ICT). The Lagrangian analysis tool LAGRANTO (Wernli and Davies, 1997) allows the identification of air mass source regions. It has been applied to study SI (e.g., Cristofanelli et al., 2003; Gerasopoulos et al., 2005) and ICT (e.g., Guerova et al., 2006; Li et al., 2005) events. The Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005), which simulates the transport and dispersion of tracers by calculating the trajectories of a multitude of particles parameterizing turbulence and deep convection, has also been used in previous studies to investigate SI (e.g., Stohl et al., 2000) and ICT (e.g., Huntrieser et al., 2005). FLEXPART was validated with data from three large-scale tracer experiments in North America and Europe (Stohl et al., 1998), and it has been used previously in many case studies of trace gas transport (Stohl and Trickl, 1999; Forster et al., 2004; Spichtinger et al., 2001), in a climatology of ICT (Stohl et al., 2002a; Eckhardt et al., 2003) and in simulating SI (Cristofanelli et al., 2003; James et al., 2003a,b).

A comparison between FLEXPART and LAGRANTO has been discussed in previous studies on stratosphere-to-troposphere exchange (STE) flux (e.g., Cristofanelli et al., 2003; Bourqui, 2006). Bourqui (2006) found that purely trajectory-based approaches yield lower-bound estimates for STE flux because they capture the large-scale transport but neither small-scale turbulent nor diffusive processes. In contrast, particle dispersion models with parameterized turbulence led to slightly larger estimates. Nevertheless, it is claimed that both models in Bourqui's study accurately identified regions subject to STE and provided realistic quantitative estimates.

To our knowledge, a statistical inter-comparison of the performance of FLEXPART and LAGRANTO on representing SI and ICT events has not been documented up to now. In this study, FLEXPART and LAGRANTO were run for the year 2005. We evaluate the models' performance on capturing SI and ICT events by comparing the model simulations with measurements at the Jungfraujoch (JFJ) site. Moreover, an intercomparison between FLEXPART and LAGRANTO is discussed. It needs to be clarified that in this paper we discuss ICT only from the North American planetary boundary layer (PBL), which brings air pollutants contributing considerably to the European O_3 budget (Auvray and Bey, 2005).

In Sect. 2 we describe the data set and model set up. In Sect. 3, we compare model simulations with measurements using two case studies. Section 4 presents a systematic comparison between models and measurements from a statistical point of view. The sensitivity of the comparison to variations of certain threshold values is determined in Sect. 5. Finally, a general discussion is given and conclusions are drawn in Sect. 6.

2 Data sets, model and methodology

2.1 Measurements

The JFJ station is located at 7.98° E and 46.55° N, 3585 m above sea level on the main crest of the Bernese Alps, Switzerland. Trace gas measurements at JFJ are performed within the Swiss air pollution network NABEL as part of the Global Atmosphere Watch (GAW) program of the World Meteorological Organization (WMO). Due to its elevated altitude, JFJ samples more than 50% free tropospheric air, particularly in winter (Zellweger et al., 2000). The concentrations of trace gases are measured by Swiss Federal Laboratories for Materials Testing and Research (Empa) and additional meteorological parameters are measured routinely by the Swiss Federal Office of Meteorology and Climatology (MeteoSwiss). The methods used for routine chemical trace gas measurements are described in detail by Zellweger et al. (2000). In our study, 1-hourly measurements of NO_y, CO, O_3, and relative humidity (RH) covering the entire year 2005 were used.

2.2 FLEXPART

To simulate the influence of SI and ICT at the JFJ site, FLEXPART (version 6.2) (Stohl et al., 2005) was employed, and was driven by model-level data from the European Center for Medium-Range Weather Forecasts (ECMWF) with a temporal resolution of 3 h (analyses at 00:00, 06:00, 12:00, 18:00 UTC; forecasts at 03:00, 09:00, 15:00, 21:00 UTC), and 61 vertical hybrid levels. All relevant fields were interpolated onto a latitude/longitude grid with horizontal resolution of $1^\circ \times 1^\circ$. We conducted forward simulations of long-range and synoptical-scale transport of anthropogenic CO tracers from the North American PBL and of stratospheric O_3 tracers, respectively.

Anthropogenic CO emissions of North America in $1^\circ \times 1^\circ$ resolution were taken from the EDGAR Version 3.2 inventory Fast Track 2000 (Olivier et al., 2005), which follows the EDGAR 3.2 method (Olivier and Berdowski, 2001; Olivier et al., 2002). The tracer

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behaves purely passively in the model, meaning that chemical degradation is ignored. The particles were released between the surface and 150 m above the ground level at a constant rate according to the EDGAR emission distribution for North America. The total number of the particles treated by the model was approximately 950 000. In a particular grid cell, the number of released particles was proportional to CO emissions in the cell related to the total CO emissions in North America. Since transatlantic transport takes place in the time range of one week (Li et al., 2005), in our simulation the passive CO tracer was allowed to be advected by the winds for 10 days, after which it was removed from the simulation. The lifetime of CO is in the time scale of a month or longer, and therefore the simulation does not account for background CO concentrations but only for enhancements over the CO background due to emissions during the last 10 days. The simulations started on 20 December 2004 in order to fulfill the 10 days spin-up of the model. Mixing ratios of CO were produced separately on a 3-hourly basis on a 3-D grid with horizontal resolution of $1^{\circ}\times 1^{\circ}$, and 25 vertical levels.

In this study, the 2 potential vorticity (PV) unit (PVU, 10^{-6} km^2 kg^{-1} s^{-1}) surface was used as the extratropical tropopause, which separates the chemically and dynamically very distinct stratosphere from the troposphere. This definition mimics reasonably well the thermal tropopause (Holton et al., 1995) and has the advantage of being based upon the dynamically significant conservation of PV in adiabatic, frictionless flow. To simulate SI impact with FLEXPART, a passive O_3 tracer was continuously released in the stratosphere within the model boundaries (60° W–60° E, 20° N–80° N), and was allowed to advect with ECMWF winds. This model domain is sufficiently large for simulating the intrusions discussed in our study, but it is worth noting that the defined domain will underestimate the intrusions frequency since the intrusions occurring outside this domain are missing in the resulting climatology. In order to obtain a rough indication of the validity of our simulations in this respect, we used calculated backward trajectories to estimate its restriction. According to the backward trajectories, 63% of the trajectories originating in the stratosphere crossed the tropopause within the domain. Nevertheless, Stohl et al. (2000) studied the influence of stratospheric intrusions on ozone concentrations in the alpine region using FLEXPART with an even more restricted domain (50° W–50° E). They suggested that although the restricted domain underestimated the influence of older stratospheric intrusions (>4 days), it adequately covers direct intrusions which are most likely to produce clear signatures in the measurements.

A statistical relationship between O_3 and PV ($M_{O_3}=M_{air}\times PV\times C\times 48/29$) was used to determine the stratospheric O_3 tracer concentration, where $C=60\times10^{-1}$ PVU^{-1} is the ratio between the O_3 volume mixing ratio and PV (in units of PVU) in the stratosphere (Stohl et al., 2000). The ratio 48/29 is used for the conversion of volume mixing ratio to mass mixing ratio where 48 and 29 represent the molecular mass of O_3 and mean molecular mass of ambient air, respectively. As for ICT, the simulation started on 20 December 2004 and ended on 31 December 2005. During the model run, approximately 3 mio particles were released, which were distributed proportional to the air density in accordance with the above formula. Each particle was tagged with a clock, which was set to zero when it remained in the stratosphere, and started to count once the particle entered into the troposphere. The clock was reset to zero if the particle returned to the stratosphere. If the particle stayed in the troposphere exceeding its assigned age class of 10 days, it was removed from the simulation. O_3 fields were produced every 6 h as 6-h averages at 1°×1° grid cells. The grid cells were 500 m thick between the surface and 10 km, 1000 m thick from 10 km to 15 km and 5000 m thick from 15 km to 20 km. The output O_3 fields only provide an estimation of the O_3 enhancement relative to the tropospheric background O_3 due to the transport from the stratosphere.

In order to achieve best representativeness for JFJ location, the model output of CO and O_3 was interpolated to the height of the JFJ site since it is located between two grid points in the ECMWF model and the topographic height of the JFJ site is much lower in the model than in reality.

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2.3 LAGRANTO

Ensembles of 10-day backward trajectories were calculated for every 6 h of the year 2005 using LAGRANTO. ECMWF analysis winds (with a temporal resolution of 6 h, horizontal resolution 1°×1° and 61 vertical hybrid levels) were used to drive the model. Because a single trajectory is often not a reliable estimate of an air parcel's path (due to lack of coherency of the flow), it is necessary to consider an ensemble of trajectories in order to obtain statistically meaningful results. Due to the fact that LAGRANTO does not simulate any diffusion, using trajectory ensemble is also a cost efficient way to qualitatively capture diffusion. Each ensemble consists of a reference trajectory (centered on JFJ at a mean pressure of 650 hPa, 7.98° E and 46.55° N) and 4 horizontally 0.5° displaced trajectories.

2.4 Identification of SI events

Besides having high O_3 concentrations, stratospheric air is characterized by low water vapor content. Thus low humidity levels can indicate stratospheric air in a tropospheric environment. Relative humidity (RH) is considered as the best humidity measure to detect SI. Specific humidity is not an appropriate measure because of its high variation with season, which can lead to an artificial bias in the evaluation of seasonal SI frequency (Stohl et al., 2000). Thus, in this paper RH in conjunction with O_3 was used to objectively identify SI events from the measurements. Enhancements over the 10-day running mean O_3 value ($M_{O_3}^{10}$) and 50% RH are taken as thresholds and are applied to O_3 and RH measurements, respectively. If O_3 values are continuously more than 10% above $M_{O_3}^{10}$ and RH values below 50% for a time period of at least 6 h, a measurement SI event is identified. The time span of the period indicates the sustaining time of the SI event, and is referred henceforth as T_s. The sustaining time T_s allows to classify the single events: we refer to events (either SI event or later on ICT event) with T_s>24 h as long events, those with T_s falling into the range between 12 and 24 h as medium events, and finally those with T_s≤12 h as short events.

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To identify a SI event from FLEXPART simulations, 10 ppb is used as threshold value of O_3 tracer. Accordingly, a FLEXPART SI event is identified if the O_3 tracer exceeds the threshold value. Based upon the analysis of LAGRANTO trajectories ensemble, we objectively identify a LAGRANTO SI event by the following approach. A stratospheric residence time can be defined for an ensemble of trajectories by the sum of the residence times of the single ensemble members. In this terms, a LAGRANTO identified SI event is characterized by a non-vanishing stratospheric residence time. To simplify our description, we introduce the new parameter T_r which represents the ensemble residence time. A LAGRANTO identified SI event at JFJ therefore means that T_r must be larger than zero at every time step during a time span of T_s, where T_s is the sustaining time of the model defined SI event, and again separates long, medium and short events.

2.5 Identification of ICT events

CO has a lifetime on the order of weeks to months (Seinfeld and Pandis, 1998), whereas the NO_y lifetime is on the order of several days. NO_y is removed from the troposphere by deposition. During the transport from the North American PBL over the North Atlantic Ocean to JFJ, most of the NO_y is removed in the free troposphere, whereas CO remains more or less constant, resulting in a relatively low NO_y/CO ratio and enhanced CO concentration compared to the background CO. According to Stohl et al. (2002b), the NO_y/CO ratio drops to values below 0.01 for air masses older than 4 days if the original ratio of NO_x/CO in the North America was assumed to be 0.16. Zellweger et al. (2003) suggest that the most aged air masses measured at JFJ are always accompanied with the lowest NO_y/CO ratios, if undisturbed free tropospheric condition occur. In our study, we chose the NO_y/CO ratio along with measured CO as control parameters to identify ICT from measurements.

To identify ICT events from the measurements, background CO under the undisturbed free tropospheric situation is calculated using the method of partitioning JFJ data according to different meteorological situations as described by Zellweger et al.

(2003). They studied the influence of different meteorological situations on JFJ data, and split them into ones with and without impact of local polluted air: for autumn/winter (September to February) 68% and for spring/summer (March to August) 40% of meteorological situations were undisturbed free troposphere without impact from local PBL air. In this study, we assume the same frequencies of meteorological situations and take into account that measurements are characterized by significantly enhanced NO_y values (not shown here) when under the influence of the local PBL air (Zellweger et al., 2003). Hence we obtain the free tropospheric situations through removing the measurements with the highest 32% NO_y values from September to February and the highest 60% NO_y values from March to August. Using the remaining measurements, we calculate the monthly CO mean value which then is used as background CO concentration. A measured ICT event is then defined to have a measured CO higher than the corresponding background CO concentration and NO_y/CO lower than its 30% monthly percentile.

An ICT event is identified from FLEXPART simulations if the CO tracer exceeds a threshold value of 5 ppb. Analogously, each ICT event is then marked with its sustaining time T_s. A similar approach is used to identify ICT events from the LAGRANTO trajectories ensemble. T_r in this case means the ensemble residence time within the North American PBL. An ICT event is identified from LAGRANTO trajectories if T_r is larger than zero.

3 Case studies

In order to illustrate our methodology, we first present two cases of SI and ICT, respectively, which were selected according to the measurements and using the criteria introduced in Sect. 2.

3.1 SI event on 15–20 February

Figure 1 presents the 5-day series of O_3 and RH measured at JFJ from 15 to 20 February, 2005, showing enhanced O_3 concentrations with continuously more than 10% of $M_{O_3}^{10}$ (not shown). RH significantly decreased from 17 February 06:00 UTC to 18 February 07:00 UTC, and fluctuated at low levels untill 19 February 06:00 UTC. Since the stratosphere shows much higher O_3 concentrations and is much dryer than the troposphere, this signal is indicative for an SI event. However, note that O_3 concentrations around 60 ppb as measured at JFJ during this event indicate significant mixing with tropospheric air since O_3 concentrations in the stratosphere are much higher. By means of the criterion described in Sect. 2.4, we identify a measured SI event which sustains for 29 h from 17 February 06:00 UTC to 18 February 11:00 UTC. Accordingly, the 29-h SI event is classified as a long SI event.

FLEXPART O_3 tracer values from 15 February 00:00 UTC to 20 February 00:00 UTC show peaking values during the period from 17 February 06:00 UTC to 19 February 00:00 UTC, which covers the period of the measured SI event, indicating good temporal agreement between the FLEXPART simulation and the measurements. O_3 tracer values during the peak period were significantly higher than the threshold value of 10 ppb. According to our terminology, the O_3 tracer indicates a FLEXPART long SI event because of the sustaining time T_s>24 h.

It should not be ignored that at 16 February 06:00 UTC, although the combination of O_3 increase and RH decrease did not fulfill the selected criteria for a SI event, changes of O_3 and RH were nevertheless discernible. This stratospheric signature at 16 February 06:00 UTC was also captured by the FLEXPART simulation. As seen in Fig. 1, a relatively narrow peak occurs exactly at 06:00 UTC in the O_3 tracer.

Figure 2 presents LAGRANTO backward trajectories (left) from 18 February 00:00 UTC to 18 February 18:00 UTC. We only present the trajectories which have crossed the tropopause. At least one member of the trajectories ensemble crossed the tropopause and descended to JFJ during the period, which indicates evident stratospheric origin of part of the air. According to the definition of a LAGRANTO SI event, a medium SI event was identified which persists for T_s of 18 h. Taking the different time resolutions between the measurements and model output, as well as the bias due to numerical treatment into account, the LAGRANTO SI event well matches with the measured SI event even though its shorter T_s. The right panel in Fig. 2 shows the trajectories ensemble started at 16 February 06:00 UTC. Although the reference trajectory stayed in the troposphere, two displaced trajectories had contact with the stratosphere. The T_r in the stratosphere at 16 February 06:00 UTC was 162 h, which indicated the air masses arriving at JFJ had clear stratospheric influence. Since the T_s is less than 12 h, it is identified as a short SI event. This LAGRANTO short SI event temporally coincides with the FLEXPART short SI event, as well as with considerable O_3 increase and RH decrease in the measurements. This indicates good agreement between LAGRANTO simulation and the measurements, as well as between FLEXPART and LAGRANTO simulations.

3.2 ICT event on 2–4 July 2005

Figure 3 presents a 5-day series of CO, NO_y, NO_y/CO values during the period from 1 July 00:00 UTC to 5 July 00:00 UTC, along with FLEXPART CO tracer values. CO showed an evident increase starting from 1 July 23:00 UTC while NO_y significantly decreased. From 1 July 23:00 UTC to 2 July 15:00 UTC, CO values were almost always higher than background CO and NO_y/CO ratios were found to be always lower than the 30% monthly percentile. According to the method described in Sect. 2.5, a measured ICT event with T_s of 16 h is thus identified.

Except for an offset of several hours, the FLEXPART simulation agrees well with the measurements: the CO tracer showed an evident enhancement from 2 July 03:00 UTC to 4 July 21:00 UTC, which is larger than the threshold value of 5 ppb adopted for this study. We therefore identify it as a FLEXPART ICT event, which is consistent with the ICT event deduced from the indicators measured at JFJ. It is worth to note that FLEXPART generated an ICT event with a much larger CO increase, probably because the decay of CO tracer is not integrated in the model simulation, which is however important in summer.

Figure 4 shows the LAGRANTO trajectories ensemble from 2 July 06:00 UTC to 3 July 06:00 UTC, which depicts the transport history of air masses from North America to JFJ. The trajectory ensemble indicates the air arriving at JFJ during this period originated from North America (top panel); the air parcels stayed over North America for about one week, and were transported across the North Atlantic Ocean to JFJ afterwards. The vertical distribution of the trajectories which had contact with the North American PBL (here taken as the levels below 800 hPa) during their 10-day history are shown in the bottom panel. The air masses were uplifted rapidly from the North American PBL to the middle troposphere, and were subsequently transported over the North Atlantic to JFJ within approximately 4 days. Note that the air originating from upper or middle tropospheric levels above North America first descended to the North American PBL, afterwards the air was quickly uplifted and transported across the Atlantic Ocean. The trajectories of the air masses show a coherent transport pattern, which is typical for transport in Warm Conveyor Belts (WCB) (Eckhardt et al., 2004). At every time step during the period from 1 July 06:00 UTC to 3 July 06:00 UTC, at least one member of the trajectory ensemble had contact with the North American PBL, meaning T_r in the North American PBL was above zero. Therefore, a LAGRANTO ICT event with T_s of 24 h was identified. This event well agrees with the FLEXPART and the measured ICT event despite a few hours offset.

4 One-year comparison between model simulations and measurements

4.1 General comparison

In the previous section it was shown that individual SI and ICT events, as identified by transport simulations (FLEXPART and LAGRANTO), agreed well with measurement based identifications of such events. The same approach is now applied to one year

(2005) of model simulations in order to obtain a statistical evaluation of the models' performance.

Figure 5 presents the one-year time series of measured O_3 and RH, FLEXPART O_3 tracer and LAGRANTO diagnosis of SI events in 2005. According to FLEXPART, the O_3 tracer was found to be higher than its threshold value of 10 ppb for 19% of the time in 2005. According to LAGRANTO, 18% of the time in 2005 air masses arriving at JFJ had contact with the stratosphere in their 10-day backward history. To be more specific, during 4% of the entire year, contact with the stratosphere took place within 4 days before the air masses arrived at JFJ. For the remaining 14%, the contact with the stratosphere took place more than 4 days ago. The results are listed in Table 1.

Figure 6 presents time series of measured CO, NO_y/CO ratio, FLEXPART CO tracer and LAGRANTO diagnosis of ICT events in 2005. Applying the criterion of Sect. 2 to FLEXPART and LAGRANTO, the influences of the intercontinental transport from the North American PBL on JFJ were quantified, respectively. FLEXPART yields a frequency of 13% for ICT impact at JFJ, which is well comparable to the frequency derived from LAGRANTO simulations. The results are also listed in Table 1.

4.2 Model comparison of SI and ICT events

Based upon measured O_3 and RH and according to the criterion used in Sect. 3, we identified 103 measured SI events, which include 4 long events, 19 medium events and 80 short events (see Table 2).

A quantitative assessment of the models' performance was accomplished using the following approach: if during the period of a measured SI event, at least one SI event is identified with the model simulation, we consider the model well captures the measured SI event. We call this approach "forward comparison". By means of this approach, we found that FLEXPART captured 3 of 4 long SI events, 12 of 19 medium SI events, but only 27 of 80 short SI events. Correspondingly, LAGRANTO captured 4 long SI events, 16 of 19 medium SI events and 41 of 80 short SI events. Although in total FLEXPART and LAGRANTO captured only 41% and 59% of 103 measurement SI events, they captured 65% and 87% of measured long and medium SI events, respectively. This indicates that under the current criteria both models are more capable of capturing long and medium measured SI events. In addition, it also shows that under the chosen thresholds, LAGRANTO is superior to FLEXPART for capturing measured SI events.

In the measurements 143 ICT events were identified in 2005 using NO_y/CO ratios and measured CO as thresholds (see Sect. 2.5). These ICT events were further split into three groups according to the length of T_s, as shown in Table 3. They consist of 8 long, 21 medium and 114 short events. After applying an analogue approach of forward comparison, it was found that FLEXPART captured 1 of 8 long, 13 of 21 medium and 34 of 114 ICT short events. On the other hand, LAGRANTO captured 4 of 8 long, 10 of 21 medium and 39 of 114 short ICT events. Compared to modelled and measured SI events, the corresponding agreement of ICT events is less convincing. This might partly be due to the larger uncertainties involved in determining ICT events from measured CO and NO_y/CO. Air masses originating from other polluted source regions having traveled long enough in the troposphere before arriving at JFJ, could also exhibit relatively low NO_y/CO ratios and CO enhancement. Even so, the results show that both FLEXPART and LAGRANTO are more capable on capturing long and medium ICT events than short ones except for FLEXPART on long ICT events.

4.3 Confirmation of modeled SI and ICT events by measurements

In a second step, comparison between model identified events and the measured events was conducted in a reverse way, namely "backward comparison". We firstly collected SI events identified with FLEXPART O_3 tracer values and LAGRANTO trajectory ensembles, respectively. Applying the same criteria as in Sect. 3, we found 121 FLEXPART SI events including 16 long, 21 medium and 84 short events, and 177 LAGRANTO SI events including 1 long, 20 medium and 156 short events. The detailed results are displayed in Table 4.

The approach used in backward comparison is analogue to the forward one: A model SI event is assumed to be confirmed by the measurements if at least one measured

SI event (independent of its length) is found during the period of the model SI event. Comparing all FLEXPART SI events with measurements, we found that 75% of 16 long, 62% of 21 medium and 23% of 84 short events were confirmed. Correspondingly, for LAGRANTO we found 60% of 20 medium and 34% of 156 short events confirmed by the measurements. Astonishingly, the only long event was not confirmed by the measurements. However the measured NO_y concentration during this long SI event was found to be significantly elevated, which might be due to the mixing with uplifted polluted air from the planetary boundary layer, causing increased O_3 titration, thus to large extent weakening the stratospheric signature. The results of confirmed events in percentages are shown in brackets in Table 4.

We present the results of the backward comparison on ICT events in Table 5. In order to highlight the ICT from the North American PBL, we considered only the trajectories which have contact with the North American PBL and have no contact with the European PBL. Therefore, we could to a large extent diminish the impact of European PBL NO_y-rich air on NO_y measurements, and solely focus on the impact of long-range transport from the North American PBL on air quality at JFJ. In the year 2005, 118 LAGRANTO ICT events were identified, which include 1 long, 19 medium and 98 short events. Comparison with measurements shows that 100% of long, 47% of 19 medium and 43% of 98 short events were confirmed by measurements. As for FLEXPART, applying the method described in Sect. 2.5 to FLEXPART CO tracer, we identified 80 ICT events including 14 long, 19 medium and 47 short events. In comparison with measurements, it was found that for FLEXPART, 71% long, 68% of 19 medium and 36% of 47 short events were confirmed by measurements.

5 Sensitivity study

In this section, we investigated the sensitivity of the agreement discussed in Sect. 4 between FLEXPART or LAGRANTO and the measurements to varying threshold values of the control parameters. With respect to SI identification of the measurements, the threshold value of O_3 was changed from 10% $M^{10}_{O_3}$ to 7.5% $M^{10}_{O_3}$ and 5% $M^{10}_{O_3}$, respectively. The RH threshold value was separately changed from 50% to 40%, and then to 60%. The threshold value of NO_y/CO was changed to 20% and 40% of its monthly percentile. For the SI and ICT identification based upon FLEXPART simulation, 15 ppb and 10 ppb as threshold values were additionally applied to the FLEXPART O_3 tracer and CO tracer. For LAGRANTO SI and ICT events, new residence times T_r (12 and 24 h) in the stratosphere and in the North American PBL were applied. With every alteration of the above mentioned threshold values, the calculations of Sect. 4 were repeated. The results of the agreement between models and measurements are displayed as percentages in Fig. 7 for SI events and in Fig. 8 for ICT events. In both figures, the upper panel shows the results of the foward comparions, the lower one the results of the backward comparisons.

The general pattern in Figs. 7 and 8 shows that the agreement between FLEXPART or LAGRANTO and measurements depends significantly on the applied threshold values of the control parameters. Although an apparent change on the agreement was found when changing the threshold values, the results were still in line with the conclusion drawn in Sect. 4, suggesting that both FLEXPART and LAGRANTO are well able to capture long and medium SI and ICT events, however less favorable on capturing short SI and ICT events. The figures also show that generally better agreement was achieved when using less strict threshold values. Few exceptions will be discussed in the following.

Figure 7 (top panel) shows when increasing the threshold value of O_3 from 10 to 15 ppb, the response on the agreement between FLEXPART and measurement was unchanged for long and medium SI events, meaning all measured long and medium SI events correspond to FLEXPART O_3 tracer of above 15 ppb. Moreover, when solely increasing the threshold value of T_r in the stratosphere from 0 to 12 h and then to 24 h, agreement between LAGRANTO and measurements on long SI events also showed no response to the alteration, indicating all measured long SI events correspond to large stratospheric T_r (>24 h) characterized in the trajectories ensemble. This finding suggests the measured long SI events were consistent with significant FLEXPART O_3 elevation and large T_r in the stratosphere as obtained from the analysis with LAGRANTO trajectories. Hence, there is a good agreement between FLEXPART or LAGRANTO and measurements, as well as between FLEXPART and LAGRANTO on the identification of significant long SI event.

Figure 8 (top panel) shows generally modest agreement between FLEXPART or LAGRANTO and measurements in identifying ICTs, confirming the conclusion from the statistical point of view that a combination of NO_y/CO and CO is still unable to accurately identify ICT from the North American PBL. Note that the agreement between LAGRANTO and measurement on the measured long and medium ICT events was reasonably stable when changing the threshold value of NO_y/CO from 20% to 30%, and then to 40% of its monthly percentile. Figure 8 (bottom panel) shows that the agreement between LAGRANTO and measurements was not changed if we changed either the threshold value of NO_y/CO from 20% to 30%, or 40% of its monthly percentile or increased T_r in the North American PBL from 0 to 12 h and 24 h. This suggests that LAGRANTO long ICT events were characterized with long residence times in the North American PBL (>24 h), which is consistent with significant ICT impact in the measurements as the temporal corresponding NO_y/CO ratios were always below the 20% monthly NO_y/CO percentile.

Finally, the impact of local pollution on our results was considered. As described in Sect. 3.2, 68% in winter/spring and 40% in summer/autumn of meteorological conditions indicate free troposphere at the measurement site. These rules, used to calculate the CO background, were derived for 1997/1998 and might not hold for the year under investigation due to the natural year-to-year variability. Therefore, sensitivity experiments were performed with 60%, 70% and 75% in winter/spring and 35%, 45% and 50% in summer/autumn to calculate the background CO concentraion. No significant difference was found on the results (not shown here).

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6 Conclusions

Measurements, the particle dispersion model FLEXPART and the trajectory tool LAGRANTO were used in parallel to identify stratospheric intrusion (SI) and intercontinental transport (ICT) events occurring at the high mountain measurement site Jungfraujoch (JFJ) in 2005. Our particular interest was devoted to evaluate the performance of the two models by intercomparing with the measurements. This study represents the first attempt to investigate the models' capability of representing SI and ICT events in a statistical way.

In order to identify SI events, we used a methodology based upon the analysis of control parameters: O_3 and relative humidity (RH) values recorded at JFJ, stratospheric O_3 tracer concentrations in the output of FLEXPART simulations and potential vorticity (PV) values along LAGRANTO trajectories. For the identification of ICT events, CO and NO_y/CO ratios calculated from measurements, CO tracer concentrations in the output of FLEXPART and altitude values along LAGRANTO trajectories were used as control parameters. A certain threshold value was assigned to every parameter allowing to objectively identify SI and ICT events.

Firstly, case studies of SI event around the period of 17–18 February and ICT event in the period of 2–4 July 2005 were performed from measurements and from FLEXPART and LAGRANTO simulations, respectively. The intercomparison showed that SI events identified separately from models and measurements are temporally offset by several hours of each other. The sustaining time T_s of the SI events also differed by a few hours from each other. Nevertheless, considering the different time resolutions between measurements and model output and considering the different numerical treatment of the models, both FLEXPART and LAGRANTO simulations were in good agreement with measurements for this case. At 16 February 06:00 UTC, both FLEXPART and LAGRANTO identified a short SI event, which however was not identified in the measurements due to the relatively strict thresholds applied. However, visual inspection indicates a clear, but weak SI signal in the measurements. This suggests that the intercomparison to some extent depends on the used threshold values (see below).

The main focus of the study was put on the statistical comparison between models and measurements. We conducted the comparison in forward and backward mode, respectively. In forward comparison, we identified SI and ICT events based on 1-year measurements in 2005. The identified events were then compared with FLEXPART and LAGRANTO simulations, respectively. Regarding to SI events, the comparison results show FLEXPART captured 75% of 4 long (sustaining time >24 h), 63% of 19 medium (≥12 h, and ≤24 h) and only 34% of 80 short SI events (≤12 h). LAGRANTO, on the other hand, captured 100% of 4 long, 84% of 19 medium and 51% of 80 short SI events. The results suggest that both FLEXPART and LAGRANTO are capable of capturing most of the long and medium SI events, but encounter severe difficulties in capturing short SI. It is also noteworthy that LAGRANTO performed better than FLEXPART which was probably because LAGRANTO used relatively less strict threshold values. Low agreement between models and measurements on short SI events points out the higher uncertainty of the realistic representativeness of short SI events themselves. As for ICT events in forward comparison, FLEXPART captured 13% of 8 long, 62% of 21 medium and 30% of 114 short ICT events. Correspondingly, LAGRANTO captured 50% of 8 long, 48% of 21 and 34% of 114 short ICT events. Although both models still showed better consistence with measurements on long and medium ICT events than on short events, the agreement on ICT events between either FLEXPART or LAGRANTO and measurements in general was worse compared to SI events. This is most likely due to the fact that a combination of NO_y/CO ratio and measured CO is inadequate to distinguish in particularly ICT air masses from the North American PBL.

In backward comparison, we applied the approach in the reverse order. The threshold values were applied to the model simulations, and SI and ICT events accordingly identified for the year 2005. The model identified SI and ICT events were then compared with measurements. Regarding the model defined SI events, our results show that more long and medium SI events were identified with FLEXPART than with LA-

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GRANTO. We identified 16 long and 21 medium SI events using FLEXPART's O_3 tracer, but only one long and 20 medium SI event using LAGRANTO trajectory ensemble. The difference is probably because an age class criterion of 10 days was used in the FLEXPART simulation, meaning that O_3 tracers were allowed to be transported in the troposphere at most for 10 days after crossing tropopause. On the other hand, 10 days was indeed the entire transport time of LAGRANTO trajectories, including the time within the stratosphere and troposphere. The comparison with measurement showed that about 75% of 16 FLEXPART long and 62% of 21 medium SI events were confirmed by measurements. Correspondingly, about 60% of 20 LAGRANTO medium events were confirmed by measurement. Astonishingly the single LAGRANTO long SI event was not confirmed probably due to mixing with locally uplifted polluted air, which covered the stratospheric signature. As for short SI events identified by either FLEXPART or LAGRANTO, the agreement with measurements was generally low, which manifests the considerable uncertainty when using models to investigate short SI events compared to long and medium ones. Finally, coming to model defined ICT events, the agreement between model and measurement was much better than that found in forward comparison. 71% of 14 FLEXPART long and 68% of 19 medium ICT events were confirmed by measurement. For 47 short ICT events, 36% of them were confirmed. For LAGRANTO ICT events, one long, 47% of 19 medium and 43% of 98 short events were confirmed by measurement. The results show that both FLEXPART or LAGRANTO are suitable tools to represent long and medium ICT events, and are relatively favorable for investigating short ICT events. The results reveal again the restriction of measured NO_y/CO ratios and CO to assess ICT events from the North American PBL.

We also conducted sensitivity studies to check the dependence of the agreement between models and measurements on the used threshold values. We applied different thresholds to relevant control parameters, respectively. For every alteration of some threshold value, the same approaches used in forward and backward comparisons were repeated. The results suggest that generally less strict threshold values resulted

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in higher agreements both for forward and backward comparisons. For every alteration, the percentage change of agreement were within 20% except in backward comparison for LAGRANTO long SI events, where the agreement between LAGRANTO and measurements changed tremendously from 0% to 100%. However due to the small quantity (one) of the LAGRANTO long SI events, this change did not have any statistical meaning. The sensitivity experiments confirm again that both FLEXPART and LAGRANTO were well capable of capturing long and medium SI events, however have problems on capturing short SI events.

A sensitivity study of the agreement on ICT events reveals that in forward comparison, although when adopting less strict threshold values, a higher percentage of measured ICT events were captured by the models, the agreement between measurements and models was still low. In contrast, during backward comparison, the results suggest that the agreement between models and measurements was much more improved, which further confirmed our conclusion drawn in the previous section that a combination of NO_y/CO ratio and CO is restricted to be applied to access ICT event from the North American PBL.

In summary, our analysis shows that particle dispersion and trajectory tools can be used to identify SI and ICT events. They both capture well long-lasting, significant events, but encounter problems when considering short events.

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Table 1. Frequencies of the influences of stratospheric intrusion and intercontinental transport from the North American PBL on JFJ in 2005 according to FLEXPART and LAGRANTO, respectively.

Type	SI	ICT
FLEXPART	19%	13%
LAGRANTO	18%	12%
LAGRANTO (0–4D)[a]	4%	–
LAGRANTO (4-10D)[b]	14%	–

[a] Events occurred within 0–4 days prior to arriving at JFJ.
[b] Events occurred within 4–10 days prior to arriving at JFJ.

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Table 2. Agreement (in number of events and in percentage) between FLEXPART and LAGRANTO simulations and the measured SI events at JFJ in 2005 according to residence time T_s in the stratosphere.

group	T_s	Measurements	FLEXPART	LAGRANTO
Long SI	>24	4	3 (75%)	4 (100%)
Medium SI	12–24	19	12 (63%)	16 (84%)
Short SI	≤12	80	27 (34%)	41 (51%)
Total	–	103	42 (41%)	61 (59%)

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Table 3. Agreement in percentage between FLEXPART and LAGRANTO simulations and the measured ICT events at JFJ in 2005 as a function of T_s in the North American PBL.

group	T_s	Measurements	FLEXPART	LAGRANTO
Long ICT	>24	8	1 (13%)	4 (50%)
Medium ICT	12–24	21	13 (62%)	10 (48%)
Short ICT	≤12	114	34 (30%)	39 (34%)
Total	–	143	48 (34%)	53 (37%)

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Table 4. Agreement in percentage (shown in bracket) between models identified SI events and the measurements at JFJ in 2005 as a function of T_s of the event in the Stratosphere.

group	T_s	FLEXPART	LAGRANTO
Long SI	>24	16(75%)	1(0%)
Medium SI	12–24	21(62%)	20(60%)
Short SI	≤12	84(23%)	156(34%)
Total	–	121(36%)	177(37%)

Table 5. Agreement in percentage (showing in the bracket) between FLEXPART and LAGRANTO simulations and the measured ICT events at JFJ in 2005 as the function of T_s in the North American PBL.

group	T_s	FLEXPART	LAGRANTO
Long ICT	>24	14(71%)	1(100%)
Medium ICT	12–24	19(68%)	19(47%)
Short ICT	≤12	47(36%)	98(43%)
Total	–	80(50%)	118(44%)

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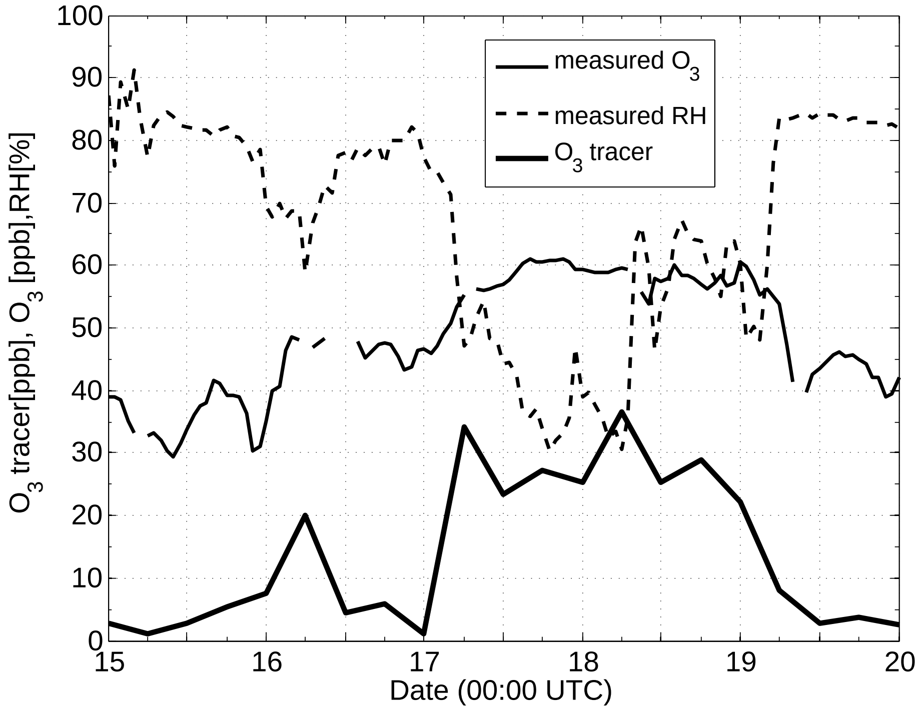

Fig. 1. Measurements and tracer model results for the period between 15 February 00:00 UTC and 20 February 00:00 UTC showing FLEXPART's stratospheric O_3 tracer (thick solid line) and the measured O_3 (thin solid line) and RH (thin dash line)

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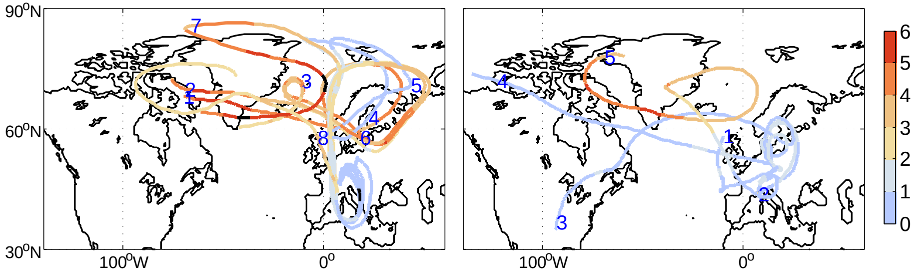

Fig. 2. Backward trajectories that crossed the tropopause during the event described in Fig. 1. Left: arrival on 18 February (at 00:00 UTC: trajectories 1, 2, 3; 06:00 UTC: trajectories 4, 5, 6; 12:00 UTC: trajectory 7; 18:00 UTC: trajectory 8). Right: Ensemble of trajectories released at 16 February 06:00 UTC (Reference trajectory 1 and displaced trajectories 2,3,4 and 5).The colorbar represents PV values.

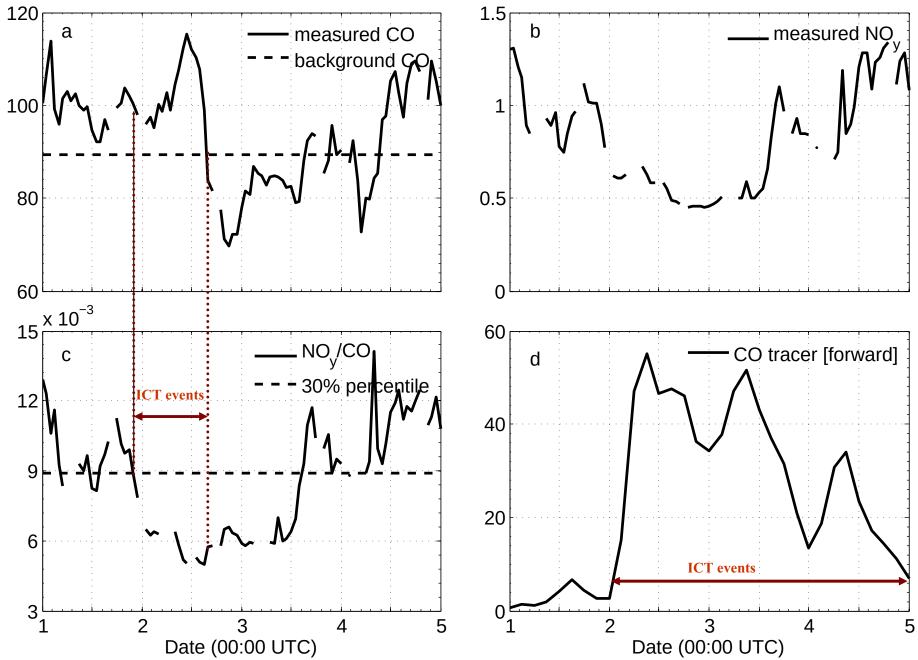

Fig. 3. Measurements and FLEXPART model results for the period from 1 July 00:00 UTC to 5 July 00:00 UTC. **(a)** measured CO and corresponding background CO; **(b)** measured NO_y; **(c)** NO_y/CO ratios; **(d)** FLEXPART simulated CO tracer in forward mode.

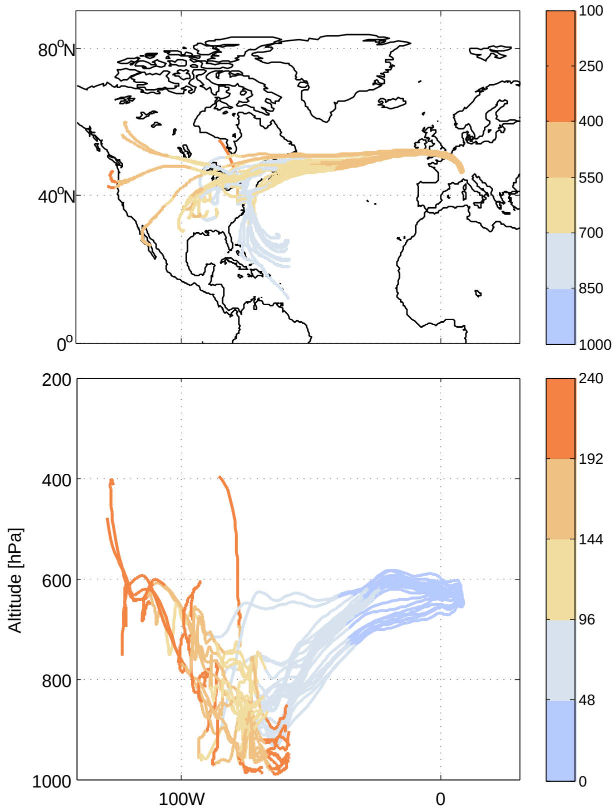

Fig. 4. Ensemble of backward trajectories starting at JFJ (top:colorbar indicates altitude in hPa) and altitude evolutions along the trajectories for the 240 h interval from 2 July 06:00 UTC to 3 July 06:00 UTC, 2005 at JFJ (bottom: colorbar indicates the time to JFJ).

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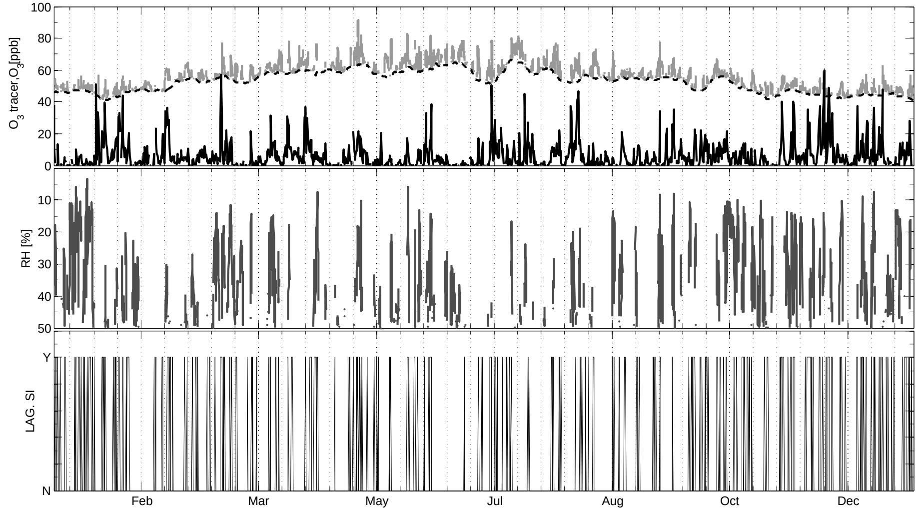

Fig. 5. Top: O_3 measurements (grey line) greater than the 10-days running mean $M^{10}_{O_3}$ (dash black line), and FLEXPART O_3 tracer mixing ratios (solid black line) in 2005. Middle: RH measurements (grey line). Bottom: LAGRANTO SI diagnosis.

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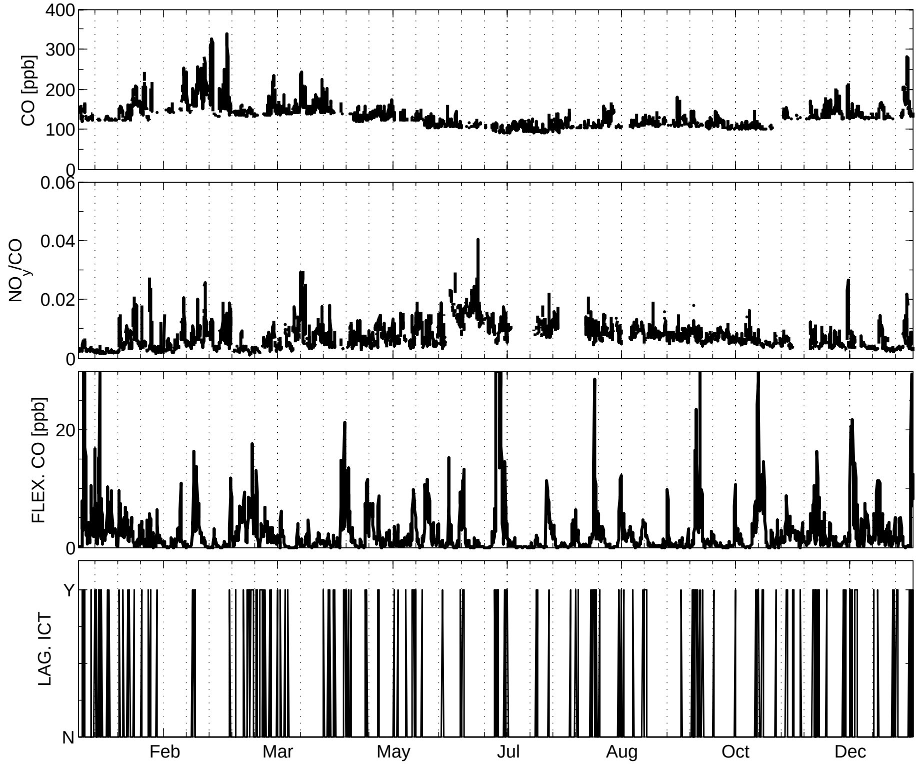

Fig. 6. Measured CO, NO_y/CO ratios, FLEXPART CO and LAGRANTO ICT diagnosis in 2005. Only measured CO which is higher than background CO is shown.

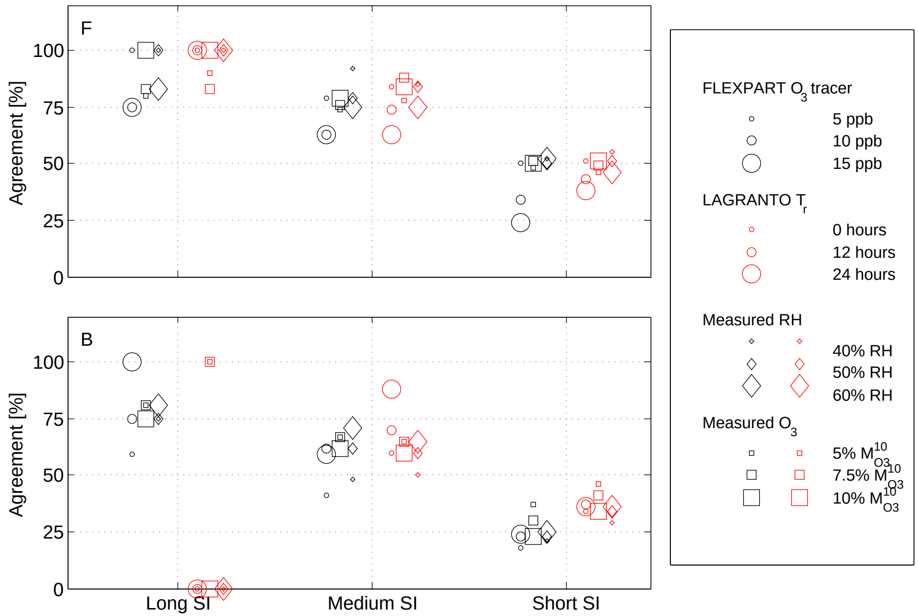

Fig. 7. Sensitivity of the agreement on SI events between FLEXPART simulation (black) or LAGRANTO simulation (red) and the measured SI events in forward comparison (top) and in backward comparison (bottom) to varying threshold values of FLEXPART O_3 tracer, LAGRANTO T_r, measured O_3 and RH.

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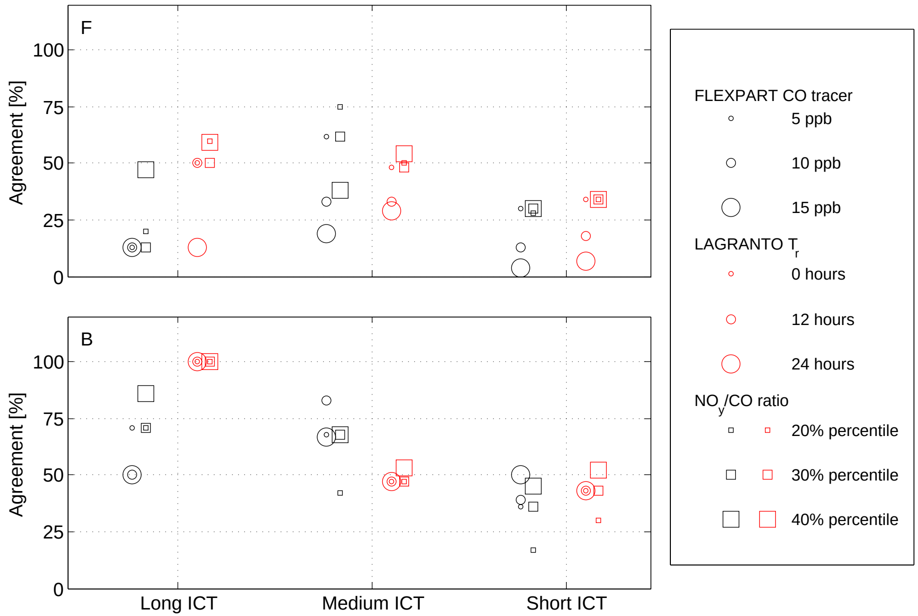

Fig. 8. Top: Sensitivity of the agreement between FLEXPART simulation (black) or LAGRANTO simulation (red) and the measured ICT events in forward comparison to varying threshold values of FLEXPART CO tracer, LAGRANTO T_r and NO_y/CO ratio. Bottom: In backward comparison.