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Tracer analysis of transport from the boundary layer to the free troposphere

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Abstract. A mesoscale meteorological model with passive tracers is applied in order to analyze transport from the atmospheric boundary layer to the free troposphere. The validation of the model against aircraft measurements indicates that the model adequately simulates the tracer distributions in the free troposphere as well as in the boundary layer. Budget calculations of passive tracers are performed in order to estimate the amount of transport in the area of a frontal system. The results show a very effective uplift associated in particular with a WCB transporting up to ~ 70% of a passive tracer initialized in the boundary layer to the free troposphere within 3 days.

1. Introduction

In order to explain the variability of chemical species in the free troposphere it is necessary to analyze not only the chemistry but also the transport of anthropogenic pollutants and to understand the dynamical processes responsible for the vertical uplifting of airmasses from the atmospheric boundary layer (ABL) to the free troposphere (FT). Three major processes are responsible for high vertical uplifting (some kilometers): frontal systems, deep convection and orographically induced vertical motions. The processes of vertical uplifting are not well understood yet and in particular poorly quantified. They are very important for the transport of pollutants to the FT where they can affect the ozone and radical budget due to e.g. the ozone production efficiency per unit NO_x, which is approximately 5–10 ppb O_3 / ppb NO_x in the continental ABL and 20–100 ppb O_3 /ppb NO_x in the FT [*Liu et al.*, 1987]. Measurements of trace gases in frontal systems gave clear evidence of strong uplifting of airmasses from the ABL to the FT associated with WCBs over western Europe, which are defined as streams of warm air which propagate along and ahead of a cold front typically ascending [*e.g. Bethan et al.*, 1998]. Some studies have already been carried out on the continental scale in order to estimate the amount of pollutant transport from the ABL to the FT [*e.g. Liang et al.*, 1998]. The most popular approach for the analysis of vertical transport is the use of trajectory models [*e.g Wernli and Davies*, 1997; *Stohl*, 2001]. However, continental scale and trajectory models tend to rely on data sets with coarse spatial and temporal resolution often insufficient to resolve mesoscale features of vertical transport (including convection) related to frontal systems.

In the present study passive tracers are transported online in a mesoscale model with high horizontal and in particular high vertical resolution in order to analyze and quantify the vertical transport associated with a frontal system. The tracer transport is validated against aircraft measurements.

2. Measurements and meteorological context

On August 5, 1999 measurements of primary pollutants, photooxidants and intermediate products including radical species were performed on board of the Met. Research Flight C–130 aircraft. In this paper we concentrate on the CO and specific humidity measurements. CO is a good tracer of continental emissions due to its lifetime of several weeks. It was measured with the resonance fluorescence method with a temporal resolution of 1 s and an accuracy of 4% [*Gerbig et al.*, 1999].

The meteorological situation at the beginning of August 1999 was characterized by a depression located west of Ireland which slowly moved south–eastward. On August 5, 1999 the associated cold front extended over England and France towards northern Spain and moved slowly eastward. The warm front extended over England and northern France while the occlusion turned over Ireland (Fig. 1).

3. Model description

The mesoscale model (Méso–NH) applied for the analysis of this flight is a non–hydrostatic anelastic equation model developed by Météo France and Centre National de la Recherche Scientifique [*Lafore et al*, 1998]. Key parameterizations used for the presented case study include a mass conserving multidimensional positive def-

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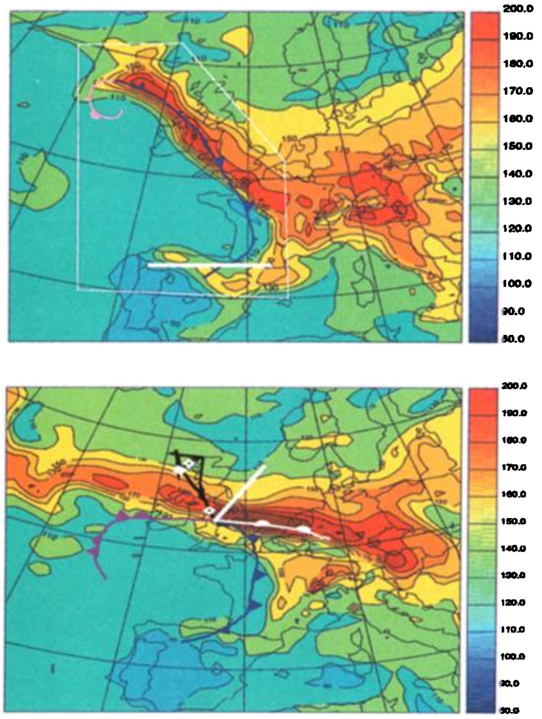

Figure 1. The CO passive tracer field [ppb] at ~ 3 km and the position of the frontal system on August 3, 1999 at 12 UTC (upper frame) and August 5, 1999 at 12 UTC (lower frame). Upper frame: The white box marks the area of the WCB (see text). Lower frame: The black line indicates the flight track, the white rectangles the positions of the CO maxima (see text).

inite transport algorithm [*Lafore et al*, 1998]. The convection parameterization is based on the *Kain and Fritsch* [1993] scheme and the turbulence scheme follows *Bougeault and Lacarrére* [1989]. The presented simulation was initialized on August 1, 1999 at 0 UTC and carried out with a horizontal resolution of 60 km on 62×62 grid points centered at 0° and 51°N (Fig. 1). In the vertical a terrain–following coordinate system is used with 71 unequally spaced levels from the surface up to 16 km. A high vertical resolution of 40–80 m is applied for the ABL and ~ 300 m for the lower and free troposphere. For initial and boundary conditions ECMWF analyses

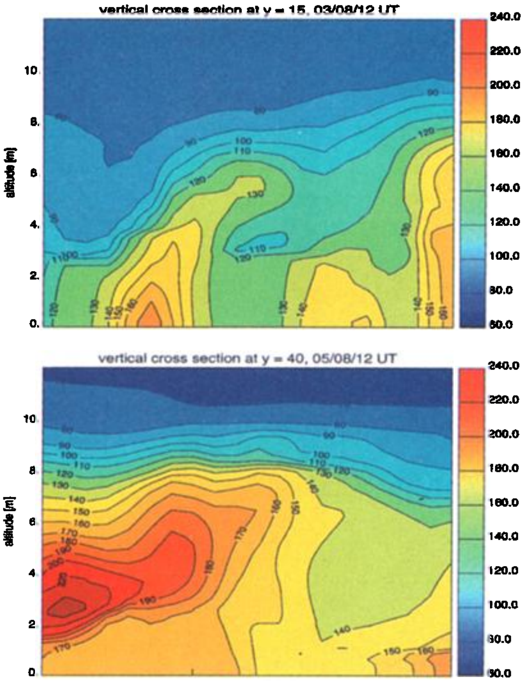

Figure 2. Vertical cross sections of the CO tracer [ppb] along the white lines marked in Fig. 1.

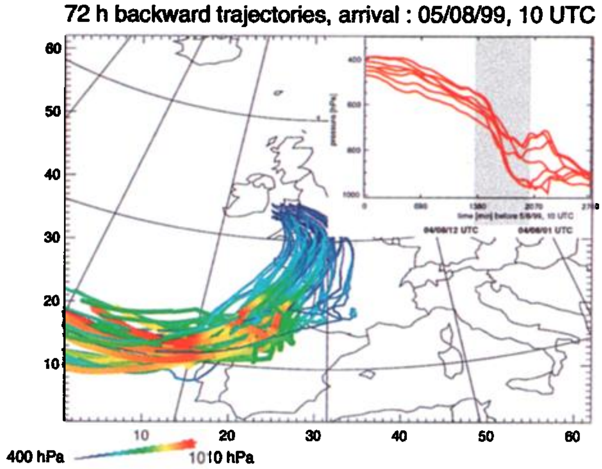

Figure 3. Ensemble of backward trajectories released on August 5, 1999 at 10 UTC in the area of the CO maximum. Large frame: horizontal displacement and pressure. Small frame: pressure as function of time. The shaded area corresponds to August 4 from 1 to 12 UTC.

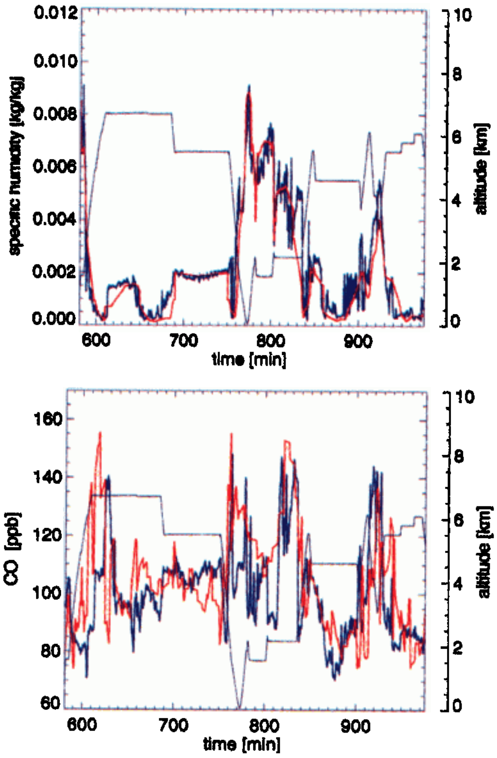

Figure 4. Comparison between specific humidity (upper frame) and CO (lower frame) measured along the flight track (blue) and specific humidity and CO passive tracer as calculated by the model (red). The black line shows the altitude of the flight.

with the spectral resolution T213 and 50 hybrid levels are used.

In order to analyze the vertical transport from the ABL to the FT a CO-like passive tracer and an ABL tracer were implemented into the Méso-NH model. The CO tracer distribution was calculated using as initial background and as boundary conditions monthly means for August 1999 calculated by the global model MOZART [*Hauglustaine et al.*, 1998] and EMEP CO emissions [*Berge et al.*, 1997] with a temporal resolution of 1 h. For the analysis of the impact of CO emissions on the FT a CO emission tracer (initialized to zero at the beginning) was transported separately. The boundary layer tracer was initialized to a constant value throughout the ABL. The ABL height was defined as the height at which the Richardson number was smaller than 0.05.

As a second tool for the analysis of transport processes and the interpretation of the measurements, three-dimensional trajectory calculations were performed using the wind fields from Méso-NH with a temporal resolution of 1 h.

4. Model results and trajectory analysis

Figure 1 shows as an example the horizontal distribution of the CO passive tracer at ~ 3 km on August 3 at 12 UTC and August 5 at 12 UTC. Figure 2 displays two vertical cross sections at the same dates. The tracer field reveals on August 3 an uplifting of western European emissions (Fig. 2, upper panel) and a northward transport ahead of and along the cold front towards central UK and the Atlantic (Fig. 1). At the same time central and eastern European emissions are transported north-westwards (Fig. 1). Ensembles of four day 3D backward trajectories terminating in the morning hours of August 5 in the FT between 6.5 and 7.5 km in the area of the southern part of the flight track all indicate rapid ascent of up to 6500 m during the previous 48 to 60 hours along and ahead of the cold front (Fig. 3). As shown by meteosat images, this ascent occurred within a cloud system extending over north-western Spain and the Bay of Biscay. As a result of this uplifting of polluted airmasses within the WCB the vertical cross section of the CO tracer reveals over central UK on August 5 a polluted layer between 1.5 and 6 km altitude with a maximum exceeding 220 ppb between 2 and 4 km (Fig. 2, lower panel).

Table 1. Percentages of the ABL passive tracer in the FT, above 4 km and above 6 km in the area of the WCB.

day / hour	FT [%]	> 4 km [%]	> 6 km [%]
3/8, 00 UTC	21.4	3.4	1.5
3/8, 12 UTC	30.4	10.3	4.7
4/8, 00 UTC	49.7	21.4	9.8
4/8, 12 UTC	70.1	45.2	26.5
5/8, 00 UTC	66.3	41.3	21.2
5/8, 12 UTC	64.2	38.9	18.4

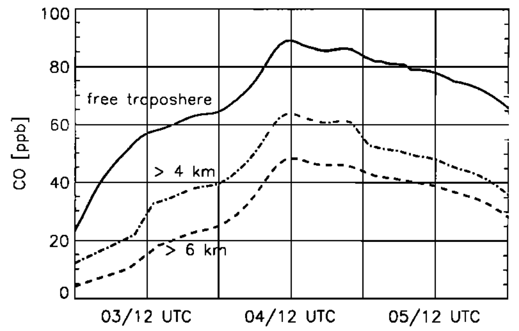

Figure 5. Temporal evolution of the mean mixing ratio of the CO emission tracer in the free troposphere from August 3, 1999 at 0 UTC to August 5, 1999 at 18 UTC.

As an example of the model validation the comparison between measured and modeled values of specific humidity and CO are displayed in Figure 4. The specific humidity shows an excellent agreement between the main features at all flight altitudes, in particular in the FT between 4 and 7 km altitude.

Four peaks of CO exceeding 140 ppb were observed at different altitudes along the flight track and well reproduced by the model (Fig. 4). In this paper we concentrate on the first maximum, which was measured north of Wales in the FT at an altitude of ~ 6.7 km (Fig. 1). The strong northeastward ascent, the meteosat pictures, the tracer flow and the position of the cold front lead to the conclusion that the WCB is the mechanism responsible for the uplifting of pollutants towards the FT over central and western UK. The three other maxima of CO were observed within the central and eastern European plume north-west of Scotland at altitudes of ~ 1.7 km, 1.3 km and 2.2 km (Fig. 1). The temporal shift of ~ 10 min between the first measured and modeled CO maximum (Fig. 4) corresponds to a horizontal distance of ~ 50 km which is smaller than one grid box and therefore within the uncertainty of the model resolution. We conclude that the spatial and temporal resolution of the model is appropriate for this case study since the model reproduces remarkably well the observed features at all flight levels. These results give an overall confidence in the model's performance in simulating meteorological parameters and tracer transport in the ABL as well as in the FT.

5. Tracer budget calculations

In order to estimate the amount of exchange between the FT and the ABL and to understand to what extent the tracer distribution in the FT is governed by dynamics independent of the spatial and temporal variation of emissions a boundary layer tracer was initialized on August 2, 1999 at 12 UTC in addition to the CO tracer. The initialisation time was chosen according to the time when the backward trajectories shown in Figure 3 reached the ABL. The tracer was transported online over a time period of 72 h. The area of the WCB

(~ 4 $\times 10^6 km^2$, Fig. 1) was determined calculating 3D trajectories [*Wernli and Davies*, 1997; *Stohl*, 2001]. It covers the region of the frontal system where the trajectories reveal an ascent from the ABL to the FT of more than 3000 m. In this area the fluxes out of the ABL were summed up and corrected by the in- and outflow of this domain, integrated since the initialisation time and compared to the initial tracer mass in the ABL (Tab. 1). The temporal evolution shows a maximum of ~ 70% on August 4 at 12 UTC. This corresponds to the time of strongest uplifting also evident from trajectory calculations (Fig. 3). About 45% of the ABL tracer are transported above 4 km, 27% above 6 km.

In order to analyze the impact of emissions on the FT budget calculations of the CO emission tracer were carried out. Figure 5 shows the mean mixing ratio of the CO emission tracer in the FT in the area of the WCB as function of simulation time. The strongest increase occurred from August 3 at 0 UTC to August 4 at 12 UTC due to the upward and northward transport of the Spanish emissions, the northwestward transport of the French emissions and the upward transport of the British emissions. These calculations result in an increase in the mean concentration of the CO emission tracer in the FT from an initial value of 0 ppb to a maximum of of ~ 90 ppb on August 4 at 12 UTC. Above 6 km the emission tracer reaches a maximum mean concentration of 50 ppb, which amounts to ~ 50%–70% of the background CO concentration.

6. Conclusions and perspectives

In this work we present a first attempt of evaluation of vertical tracer transport in a mesoscale model associated with a frontal system. The CO and humidity structures observed along the flight track are well reproduced by the model allowing to use the passive tracer for budget studies. The results show that vertical uplifting of boundary layer air is very important for the budget of chemical species in the FT. In the presented case study up to ~ 70% of the ABL tracer are transported from the ABL to the FT in the area of the WCB. As a result of this uplifting the CO mixing ratio increases in the FT by ~ 70%–100% in this case study simulation.

A question which deserves further investigation concerns the representativity of this study. It is necessary to analyze more cases in order to achieve statistically significant results. This work opens the possibility to use mesoscale models as a benchmark for the validation of larger scale models (GCMs) which then in turn could be used for climatological studies.

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