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Abstract

During the Mediterranean Intensive Oxidant Study (MINOS) in August 2001 a total of 14 measurement flights were performed with the DLR Falcon aircraft from Heraklion, Crete. One objective of this campaign was to investigate the role of long-range transport of pollutants into the Mediterranean area. An analysis of 5-day back trajectories indicates that in the lower troposphere (0–4 km) air masses originated from eastern and western Europe, in the mid-troposphere (4–8 km) from the Atlantic Ocean region and in the upper troposphere (8–14 km) from North Atlantic Ocean/North America (NAONA) as well as South Asia. We allocated all back trajectories to clusters based on their ending height and source region. The mixing ratios of ozone, nitrogen oxide, total reactive oxidized nitrogen (NO_y), formaldehyde, methanol, acetonitrile, acetone, peroxyacetyl nitrate (PAN), carbon dioxide, carbon monoxide and methane measured along the flight tracks are examined in relation to the different cluster trajectories. In the lower troposphere the mean gas mixing ratios of the eastern Europe cluster trajectories were significantly higher than that from western Europe. Considering 2-day instead of 5-day trajectories the relative differences between the concentrations of these two clusters increased. In the upper troposphere relatively high concentrations of O_3 and NO_y, combined with low CO of the NAONA trajectories indicate mixing with stratospheric air masses.

1. Introduction

A large number of chemical measurements was made on the DLR Falcon aircraft during the Mediterranean Intensive Oxidant Study (MINOS) flights in August 2001. The measurements were designed to explore the role of long-range transports of pollutants into the Mediterranean basin. As a result of the regional meteorological conditions (cloud-free, high temperatures and intense solar radiation), photochemistry can be very active (Kouvarakis et al., 2000), and the area is sensitive to air pollution. An

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overview is given by Lelieveld et al. (2002).

Backward trajectory analysis has become a widely used method for interpretation of trace gas measurements in relation to large scale air mass transports (Moody et al., 1995). The trajectories can be categorized in particular clusters to characterize different types of air masses. In most cases, the origin of the air masses and their transport routes have been used as the main criteria for clustering. By assuming that the chemical composition changes at the same rate along all transport paths, the variability in the measured trace gas concentrations can be explained in terms of the trajectory origin and emission influences along the route (Pochanart et al., 2001).

It should be mentioned, however, that the implicit assumption that the trajectories accurately represent transport pathways may sometimes be incorrect. There are many uncertainties involved in the measurements of the meteorological conditions, the generation of reanalysis data with help of the observed parameters and assimilation methods, the interpolation of the wind in time and space, and also the representation of small-scale effects (e.g. convection) in the meteorological data. Stohl (1998) estimates the accuracy of computed trajectories with position errors up to 20% of the travel distance. Nevertheless, trajectories provide a useful tool to characterize patterns in the large-scale motion.

In the next section a short overview of the measurement technique on board the Falcon is given. In Sect. 3 a general overview of the large-scale circulation over the Mediterranean area is presented. Section 4 subsequently describes the model used, and Sect. 5 presents the three dimensional trajectory analysis and the chemical characteristics of the different trajectory clusters. Section 6 concludes by summarizing the relative importance of different source regions.

2. Measurement techniques

CO, CO_2 , CH_4 , CH_3OH , CH_3COCH_3 , CH_3CN , PAN and HCHO were measured by the Max Planck Institute for Chemistry, Mainz, Germany.

For CO, CO_2 and CH_4 a Tunable Diode Laser Absorption Spectrometer (TDLAS), with a time resolution of 6 s, was used. The precision was 1.5 ppm for CO_2 , 1.5 ppbv for CO and 16.5 ppb for CH_4 and the average accuracy 1% for all species. A detailed description of the instrument can be found in Wienhold et al. (1998).

The mixing ratios of CH_3OH , CH_3CN , CH_3COCH_3 and PAN were measured using proton-transfer reaction mass spectrometry (PTR-MS). The accuracy for CH_3OH was 20%, for CH_3CN and CH_3COCH_3 15%. The measurement technique is described in detail in Lindinger et al. (1998).

The measurement of HCHO was based on the fluorimetric detection of the Hantzsch reaction product, as described by Belman (1963) or Nash (1953). The instruments noise is about 1.5% of the concentration reading up to concentrations of about 20 ppbv.

NO, NO_y and O_3 were measured by the DLR Oberpfaffenhofen and the Institute of Atmospheric Physics, Wessling, Germany. For NO and NO_y a chemiluminescence detector was used. The accuracy of the NO and NO_y measurements is 5% and 15% for concentration levels of 1 ppbv.

O_3 is measured with UV absorption technique with an accuracy of 5%. Details of the instruments are given in Schlager et al. (1997) and Ziereis et al. (2000).

3. Meteorological situation

The mean flow in the extratropics is characterized by the westerlies. The geographical position of the Azores High and the Icelandic Low can modify the zonal flow and therefore have a great impact on the weather in Central Europe and the Mediterranean region. When the High and Low are strongly developed and therefore the pressure gradient is relatively high, relatively moist air masses from the Atlantic Ocean are transported to Europe. In the alternate case, the zonal air flow is weak and meridional and blocking weather systems over Central Europe prevail. The interannually varying pressure gradient between the Icelandic Low and Azores High is generally known as the

North Atlantic Oscillation (NAO). It is linked to the general atmospheric and the oceanic circulation systems, the latter being affected by the formation of cold bottom waters in the Arctic ocean and the influx of salty water from the Mediterranean Sea through the the Strait of Gibraltar (Bolle, 2003). By definition, a relatively high pressure gradient between the Azores High and the Icelandic Low yields to a positive NAO index, a low gradient to a negative NAO index. Although its influence has been studied primarily for the winter season, NAO displays considerable monthly and interannually variability and effects have been identified for all seasons. During August 2001, the mean NAO index was 0.3 (source ftp://ftp.ncep.noaa.gov/pub/cpc/wd52dg/data/indices/tele_index.nh).

In summer the Innertropical Convergence Zone (ITCZ) and the Azores High shift toward higher latitudes. The enhanced influence of this anticylcone leads in the lower atmosphere to northerly flow toward the Mediterranean area. Depending on the geographic position of the Azores High or other quasi-stationary pressure systems over Europe, air from either the northwest or northeast is transported into the Mediterranean basin. During these months the area is directly under the downward branch of the Hadley circulation, which is driven by deep convection in the ITCZ. In the upper troposphere, poleward moving air from the ITCZ is deflected by the Coriolis force. The resulting eastward flow reaches a maximum strength in the subtropical jetstream at about 40° in summer, north of the Mediterranean. As a result of subsidence, the region is characterized by cloud-free conditions with high solar radiation intensity. Over land, convection can develop, which is generally not very deep at coastal locations. The air near the surface returns to the equatorial region in the trade winds, gathering moisture over the sea.

As a result of the northward displacement of the ITCZ the Asian Low becomes part of it, generating the Indian monsoon, with intense deep convection. For reasons of continuity, an upper-level high pressure system over the Asian surface Low is formed, the Tibetan High. The Tropical Easterly Jet (TEJ) stream is an inherent feature of the Indian summer monsoon. It is a belt of strong easterly winds, which is a part of the southern periphery of the upper tropospheric anticyclone. This TEJ between 200 and

100 hPa transports air from Asia over North Africa and, aided by the upper tropospheric anticyclone over the Arabian Peninsula, toward the eastern Mediterranean. Under the influence of the subtropical jetstream and the westerlies the air is subsequently transported back east over the Asian continent.

In summer teleconnections are found between the Mediterranean region and the South-Asian monsoon and Sahel rainfall regimes on an interannual time-scale (Ward, 1992). The sea level pressure over the eastern Mediterranean is anticorrelated with that in the Indian monsoon, mainly in the July-September period, while that in the western Mediterranean is positively correlated with a maximum during September-November. The meridional wind component is anticorrelated with the Indian monsoon over the central and eastern Mediterranean basin. This means that a more active monsoon is connected with lower sea level pressure over the eastern basin and higher over the western basin, and stronger northerly wind over most of the Mediterranean.

4. Modeling tools

4.1. Backward trajectories

The trajectories are computed with the trajectory model FLEXTRA (Stohl et al., 1995; Stohl and Seibert, 1998). Stohl and Seibert (1998), studying potential vorticity conservation, have shown that three-dimensional trajectories are more accurate than all other types, including isentropic trajectories. Thus three-dimensional trajectories were used throughout this study. In addition to the trajectory positions, FLEXTRA also provides other information, such as potential vorticity (PV), which is interpolated from grid point values. The positions of all parcels are given every 30 min.

As input data, three-dimensional wind fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) analyses are used. For this study a horizontal resolution of 2.0° and a nested domain (latitude range: 15° to 75°, longitude range: −30° to 70°) of 0.5° were employed. It has to be mentioned that the computed position

of the trajectories, especially of those trajectories that have a temporal range of several days, is dependent on the resolution of the wind data. This results from the fact that the wind fields are archived every 6 h and consequently this data has to be interpolated in time and in space. The calculations are sensitive to initial conditions in the sense that a trajectory can separate rapidly from another trajectory starting nearby (in time or space). As a consequence, uncertainties in the origin of 5-day back trajectories can add up to several hundreds of kilometers, even using the latest high-resolution global meteorological analyses (Stohl, 1998).

To determine the source regions of the air masses which were sampled during the MINOS campaign, 5-day backward-trajectories have been computed. As starting point of these trajectories the position of the Falcon aircraft was chosen along the flight track with a temporal resolution of one minute.

We divided the atmosphere into four different height intervals: level one from the ground up to 2 km, level two from 2 to 4 km, level 3 from 4 to 8 km and level 4 from 8 to about 14 km. These levels refer to the flight altitude of the Falcon aircraft during the measurements, i.e. the starting points of the back trajectories. That implies that e.g. all trajectories with ending points between 4 and 8 km are attributed to one height level regardless of the starting height 5 days earlier.

4.2. Mean trace gas mixing ratios

The temporal resolution of the measurement of the different gases was not unique. To compute mean concentrations for the trajectory clusters, the measured values have been converted to one minute averages. Hence the gas concentrations could be assigned to the corresponding trajectories, which also have a temporal resolution of one minute.

5. Source regions

On first inspection of all back trajectories computed for the MINOS campaign period we identified four main clusters of trajectories. Therefore we defined each of these clusters as representing a source region. These are: 1) western Europe; 2) eastern Europe; 3) the North Atlantic Region and North America; and 4) South Asia. These regions are depicted in Fig. 1. It should be mentioned that especially for the middle and upper tropospheric trajectories the expression ”source region” is debatable. Often the trace gases in these air parcels do not have their origin in the particular region depending on whether the trace gas life time exceeds that of the trajectory. Some trace gases are moreover chemically formed from source gases that are emitted elsewhere. In this study we nevertheless adopted this expression to distinguish the different clusters. For the approximate lifetimes of the sampled trace gases see Table 1.

By definition, if an air parcel trajectory resides over a defined region it is added to the particular cluster. Often it appears that the back trajectories reside over two or more defined regions. In this case the residence time of the air parcel above the regions is the decisive point. In this study a minimum residence time of 2.75 days is used to assign such trajectories to their source region. It is thus assumed that during this period the air mass has adopted the chemical characteristics of this region. If a trajectory does not fulfil this criterion for any region it is declared as “non-defined” and is not further considered. 184 of the 2690 computed back trajectories (6.8%) were therefore disregarded.

5.1. Ground – 2 km

Using the criteria described above we distinguish air masses on the one hand from western Europe, notably France, Germany and northern Italy, and on the other hand from eastern Europe. These two clusters are shown in Fig. 2a and 2b. 176 trajectories were counted for the western region. That is about 22% of all trajectories at this height level. 595 trajectories (about 74%) from the eastern area were registered. The

remaining 4% was ignored as being "non-defined". The mean gas concentrations of the eastern and western Europe trajectories are listed in Table 2.

It can be seen that all trajectories with source regions in eastern Europe are associated with higher concentrations than those from westerly directions, except for CO_2. In particular the mean value of PAN is nearly twice as high (609±225 to 309±94 pptv). The values of acetone (2541±296 to 1914±279 pptv), acetonitrile (194±30 to 130±24 pptv) and especially methanol (2857±792 to 1449±439 pptv) also are significantly higher in eastern European air.

From Figs. 2a and 2b we see that most of the westerly trajectories have their source 5 days back in the North Atlantic Region. Some eastern Europe trajectories move for a short time over the western European region, presumably picking up westerly pollution. To make a more exact distinction between air with typical characteristics for these two clusters we also analyzed 2-day back trajectories that resided over western and eastern Europe 99% of the time. The result is qualitatively similar as for the 5-day back trajectories, however, the mean trace gas concentration differences are even higher (Table 3). Whereas most eastern concentrations are somewhat higher, most of the western concentrations are lower, suggesting that there is additional pollution at this height level from the North Atlantic Region, possibly with a source in North America.

Acetonitrile can be used as a unique tracer for biomass burning (Holzinger et al., 1999). Accordingly the mean gas concentrations suggest more biomass burning in eastern compared to western Europe during the MINOS campaign. Figure 3 shows a map with the registered nighttime fires in Europe in August, 2001. In western Europe no fires have been registered for this period, while many occurred in eastern Europe. Many of the computed trajectories of the eastern Europe cluster moved over the region with a high density of fires north of the Black Sea (see Fig. 2b), explaining the enhanced concentration of biomass burning tracers.

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5.2. 2–4 km

The situation for the height interval between 2 and 4 km is very similar to the first level. There are also two main regions from where the air was transported toward the Mediterranean area, one from westerly and one from easterly directions. From the west 75 trajectories (23%) were counted, from the east 201 (62%). The remaining 15% was associated with other source regions. Figures 2c and 2d show the associated 5-day back trajectories. It is notable that the eastern trajectories at this height level 5 days previously do not originate as far in north as at the height level below. This is due to the different temporal variation of the air parcels' height at these two levels (not shown). In this case they originate especially over Turkey, Greece, Bulgaria and the Ukraine. Nevertheless the difference in the mean gas concentrations (see Table 4) of these western and eastern trajectories is similar to level 1. With the exception of CO_2 and NO the concentrations for the eastern Europe trajectories are higher.

For the aforementioned reasons we additionally analyzed the 2-day back trajectories. The results are listed in Table 5, showing that the mean concentrations for the eastern European trajectories change only slightly. Already nearly all 5-day back trajectories reside over eastern Europe.

In contrast, the values for the western European trajectories change. Whereas most concentrations decrease, those of short-lived gases such as NO and HCHO increase. Sources of these trace gases include in particular fossil fuel combustion (e.g. road transport). Many 5-day back trajectories start over the North Atlantic (see Fig. 2d). Mixing might occur there with other air masses which have a higher level of pollution, especially for gases with longer lifetimes. The 2-day trajectories stay relatively longer over western Europe (not shown) and consequently over regions with fresh emissions. Also for this height interval we obtain differences in the mean gas concentrations between western and eastern Europe air masses. As in the height level below the mean gas concentrations of the eastern Europe trajectories are higher, with the exception of CH_4 (west: 1859±26 ppbv, east: 1845±23 ppbv), CO_2 (west: 361±3 ppmv, east:

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361±4 ppmv) and NO_y (west: 0.9±0.3 ppbv, east: 0.9±0.2 ppbv). Only the mean NO concentration is much higher for the western Europe air masses (west: 0.08±0.01 ppbv, east: 0.02±0.01 ppbv).

As mentioned in the previous section the enhanced mean concentration of the biomass burning tracer acetonitrile, as well as methanol, acetone and CO, can be traced to the large number of fires north of the Black Sea.

It has to be emphasized, however, that the number of measurements associated with the 2-day back trajectories is very limited (between 10 and 16). Therefore extreme outliers have a large influence and the statistics for the two day trajectories is not very convincing.

5.3. 4–8 km

A large fraction, i.e. 406 of the total number of 509 computed trajectories to the 4–8 km altitude range, originates from the North Atlantic/North America region (80%). The remaining 20% is associated with other source regions. Because the number of trajectories from these regions is very low, we did not compare trace gas concentrations of different clusters at this height. Nevertheless, in Table 6 the mean concentrations for the trace gas concentrations of the NAONA cluster are listed to compare these with the NAONA region values of the height interval aloft.

5.4. 8–14 km

At this height interval two main directions appear from where the air is transported toward the Mediterranean area. Most of the air originates from the west. For these trajectories (see Fig. 2e), there seems to be a coherent cluster at the west of Africa. By computing these back trajectories for more than 5 days it can be shown that they also have their source region over North America. A separate analysis of the gas concentrations of this cluster moreover shows no significant differences with the rest of the trajectories shown in Fig. 2e. Therefore all these trajectories are treated as one cluster.

On the other hand, Fig. 2f consistently shows 5-day back trajectories originating over South Asia. Consequently these two very different clusters can be analyzed and compared. The number of the North Atlantic Ocean/North America trajectories (NAONA) is 645, which is 61% of all trajectories at this height level. For the Asian cluster 271 trajectories were counted (26%). 13% were associated with other source regions.

The mean gas concentrations for these two clusters are listed in Table 7. It appears that the standard deviations of the NAONA cluster trace gas concentrations are partly much higher than that from South Asia, notably of acetone, methanole and PAN, signifying a much higher variability of the sources in this region, associated with a much higher geographical spread of the NAONA trajectories.

In contrast to the lower height levels, neither of these two clusters is clearly characterized by enhanced mean concentrations of all trace gases. Whereas NO, NO_y, O_3 , CH_3OH and CH_3COCH_3 of the NAONA sector are higher, the values of HCHO, CO and CH_4 are higher for the South Asian cluster. The combination of relatively high O_3 and NO_y, and low CO for the NAONA cluster may partly be associated to mixing with stratospheric air masses. The North American trajectories start 5 days back at heights above 8 to 14 km, decreasing with time due to subsidence over the Mediterranean. The geographical position of many trajectories is further north, close to the jet stream, so there may be an enhanced possibility of mixing with stratospheric air. The height of the Asian trajectories, starting between 9 and 13 km, is furthermore decreasing with time associated with subsidence deeper into the troposphere. The position of these trajectories is more toward the south near the ITCZ where the tropopause is relatively high (about 16 km). Figure 4 supports the assumption of mixing with stratospheric air, showing the PV values of NAONA trajectories. To provide a better overview only a representative selection of all cluster trajectories is plotted. It can be seen that some air parcels have potential vorticity values higher than 2 PVU, being an indicator of stratospheric air masses.

Mean CO mixing ratios exceeding 80 ppbv in the middle and upper troposphere suggest a direct continental influence (Board et al., 1999). CH_4 is also enhanced

in the South Asian cluster, possibly from rice cultivation. Therefore, it is likely that South Asian air pollution is transported through the Asian monsoon convection to the upper troposphere, after witch it follows the upper-level anticyclone toward the eastern Mediterranean. The 20-day forward trajectories with starting points in two West Indian cities (Madras, Patna) in Fig. 5 supports this assumption. Figures 5a and 5b clearly show the anticylonic circlulation, while in Figs. 5c and 5d the fast upward transport of the trajectories up to about 15 km in the monsoon of convection is depicted.

6. Conclusions

We have analyzed back trajectories for the MINOS campaign in August 2001 combined with measured trace gas concentrations. The goal was to find a relationship between the origin of trajectories and the observed concentration at the time of their arrival. Other articles in the Atm. Chem. Phys. special issue of MINOS will discuss additional details of trace gas sources and their chemical influences.

As determined by the large-scale atmospheric circulation, we identified ”typical” flow patterns at different height levels. Between the surface and 2 km altitude we infer strong influences from western and eastern Europe. The same applies to the height level 2 to 4 km. About 74% of the computed trajectories at the lowest level was associated with the eastern European cluster, whereas 22% was western European. The remaining 4% was ignored as being “non-defined”. At the height 2 to 4 km these values changed to about 62%, 23% and 15%, respectively. Whereas the air between 4 and 8 km mainly originated from the North Atlantic Region (80%), there was additional strong influence between 8 and 14 km from South Asia. At this level about 61% of the trajectories came from the west, 26% from the Asian cluster. The remaining 13% was associated with other source regions.

In spite that the definition of the source regions and also associations to these regions is somewhat subjective and therefore open to discussion, substantial differences

in the mean gas concentrations between the different clusters and hence source regions are evident.

This is especially true for the lowest two height levels. The trace gas concentrations with the eastern European 5-day back trajectories were significantly higher than with the western European trajectories, signifying a higher level of air pollution. Considering 2-day back trajectories, with the additional criterion of having been 99% of this time over the defined source regions, the differences between the concentrations of eastern and western Europe air were even larger.

Generally speaking the concentrations of the trace gases at the highest level (between 8 and 14 km), originating from the NAONA region and from South Asia, do not differ as much as at the levels below, indicating a relatively higher level of uniformity in upper tropospheric air pollution in the northern hemisphere. Nevertheless these air masses did show typical chemical characteristics. Air from the NAONA source region seems to be influenced more strongly by the stratosphere. The air from South Asia did not show these influences. Since CO and CH_4 were enhanced in these masses, we infer relatively rapid transport from the surface by strong convection in the Indian monsoon.

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Table 1. Approximate lifetime of the sampled trace gases during summer

HCHO	hours
NO_x	days
NO_y	days to weeks
O_3	days to weeks
CH_3COCH_3	weeks to months
PAN	weeks to months
CO	weeks to months
CH_3OH	months
CH_3CN	months
CH_4	years
CO_2	years

Table 2. Comparison of mean concentrations for height level 1 (0–2 km) for Western Europe and Eastern Europe trajectories

	Western Europe		Eastern Europe	
	Mean	Std Dev	Mean	Std Dev
NO	0.04	0.01	0.05	0.02
NO_y	1.1	0.5	1.4	0.4
HCHO	0.9	0.2	1.3	0.4
O_3	58	5	70	6
CH_3OH	1448	439	2857	792
CH_3CN	130	24	194	30
CH_3COCH_3	1914	279	2541	296
PAN	309	94	609	225
CO_2	359	3	358	3
CO	126	17	156	23
CH_4	1871	21	1880	29

NO, NO_y, HCHO, O_3, CO, CH_4 in ppbv, CO_2 in ppmv, CH_3OH, CH_3CN, CH_3COCH_3, PAN in pptv.

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Table 3. Same as Table 2, but under consideration of 2-day back trajectories

	Western Europe		Eastern Europe	
	Mean	Std Dev	Mean	Std Dev
NO	0.03	0.02	0.05	0.02
NO_y	1.1	0.3	1.4	0.4
HCHO	0.8	0.2	1.4	0.4
O_3	58	6	69	5
CH_3OH	1461	489	3015	739
CH_3CN	122	24	198	30
CH_3COCH_3	1668	166	2577	298
PAN	320	121	627	224
CO_2	360	2	358	3
CO	113	11	158	22
CH_4	1866	22	1881	29

NO, NO_y, HCHO, O_3, CO, CH_4 in ppbv, CO_2 in ppmv, CH_3OH, CH_3CN, CH_3COCH_3, PAN in pptv.

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Table 4. Same as Table 2, but for height level 2 (2–4 km)

	Western Europe		Eastern Europe	
	Mean	Std Dev	Mean	Std Dev
NO	0.04	0.02	0.02	0.01
NO_y	0.7	0.3	0.9	0.2
HCHO	0.4	0.2	0.7	0.2
O_3	72	14	75	8
CH_3OH	1434	526	2281	559
CH_3CN	164	22	174	21
CH_3COCH_3	1658	376	2186	478
PAN	541	128	770	137
CO_2	361	3	361	4
CO	99	11	113	14
CH_4	1838	17	1842	21

NO, NO_y, HCHO, O_3, CO, CH_4 in ppbv, CO_2 in ppmv, CH_3OH, CH_3CN, CH_3COCH_3, PAN in pptv.

Table 5. Same as Table 4, however, for 2-day back trajectories

	Western Europe		Eastern Europe	
	Mean	Std Dev	Mean	Std Dev
NO	0.08	0.01	0.02	0.01
NO_y	0.9	0.3	0.9	0.2
HCHO	0.6	0.4	0.7	0.2
O_3	62	8	76	8
CH_3OH	1143	383	2257	607
CH_3CN	147	14	181	28
CH_3COCH_3	1294	276	2157	443
PAN	374	142	759	138
CO_2	361	3	361	4
CO	89	14	115	15
CH_4	1859	27	1845	23

NO, NO_y, HCHO, O_3, CO, CH_4 in ppbv, CO_2 in ppmv, CH_3OH, CH_3CN, CH_3COCH_3, PAN in pptv.

Table 6. Mean concentrations of the North Atlantic Region between 4 and 8 km

	Mean	Std Dev
NO	0.03	0.02
NO_y	0.6	0.2
HCHO	0.3	0.1
O_3	72	12
CH_3OH	985	400
CH_3CN	148	14
CH_3COCH_3	1512	320
PAN	447	146
CO_2	366	3
CO	83	11
CH_4	1833	21

NO, NO_y, HCHO, O_3, CO, CH_4 in ppbv, CO_2 in ppmv, CH_3OH, CH_3CN, CH_3COCH_3, PAN in pptv.

Table 7. Comparison of mean concentrations of North Atlantic Region (NAONA) and South Asian trajectories between 8 and 14 km

	North Atl.Region		South Asia	
	Mean	Std Dev	Mean	Std Dev
NO	0.2	0.1	0.1	0.1
NO_y	1.0	0.3	0.7	0.1
HCHO	0.3	0.1	0.9	0.3
O_3	105	31	63	9
CH_3OH	690	429	561	180
CH_3CN	146	12	147	13
CH_3COCH_3	1029	408	818	96
PAN	235	125	211	52
CO_2	369	3	365	3
CO	71	13	90	15
CH_4	1826	23	1853	24

NO, NO_y, HCHO, O_3, CO, CH_4 in ppbv, CO_2 in ppmv, CH_3OH, CH_3CN, CH_3COCH_3, PAN in pptv.

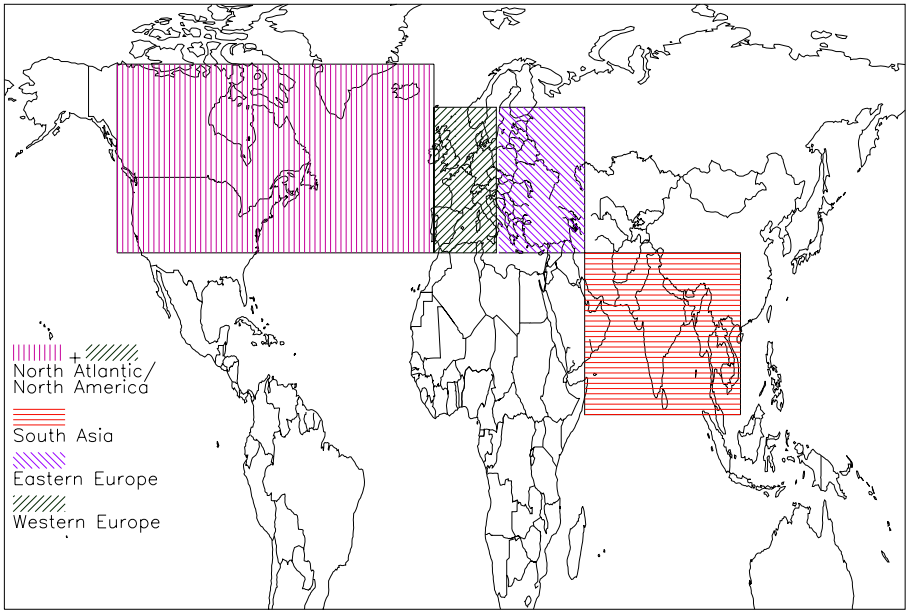

Fig. 1. Source regions of the trajectories during the MINOS campaign.

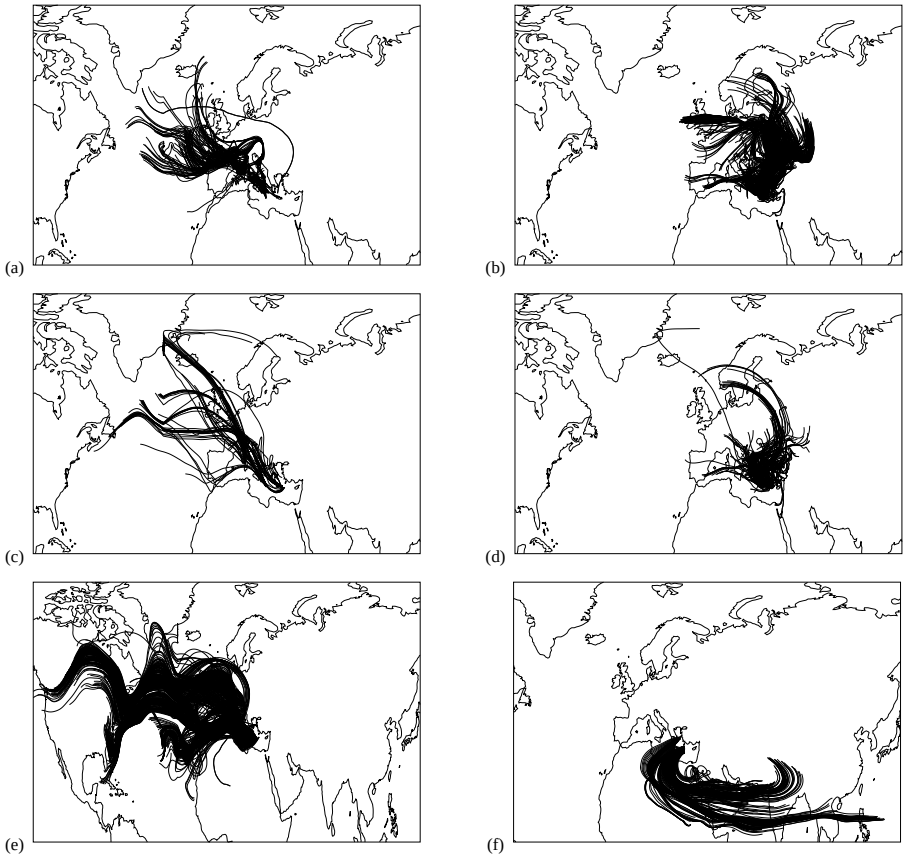

Fig. 2. Clusters of 5-day back trajectories for different height levels. **(a)** Western Europe trajectories between the ground and 2 km, **(b)** same height level for Eastern Europe trajectories, **(c)** same as (a) but between 2 and 4 km, **(d)** same as (b) but between 2 and 4 km, **(e)** North Atlantic/North American trajectories between 8 and 14 km, **(f)** same as (e) but for the South Asian cluster.

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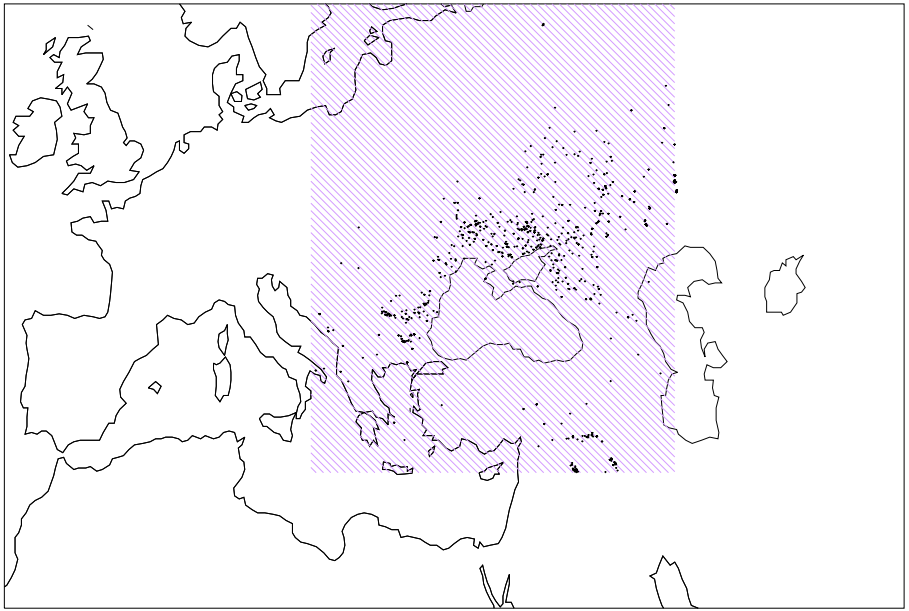

Fig. 3. Fires detected during nighttime for parts of western and eastern Europe in August, 2001. The defined region of eastern Europe is shaded. Whereas in western Europe no fire activities were registered, there were many in eastern Europe. Source: ATSR World Fire Atlas http://shark1.esrin.esa.it/ionia/FIRE/AF/ATSR/.

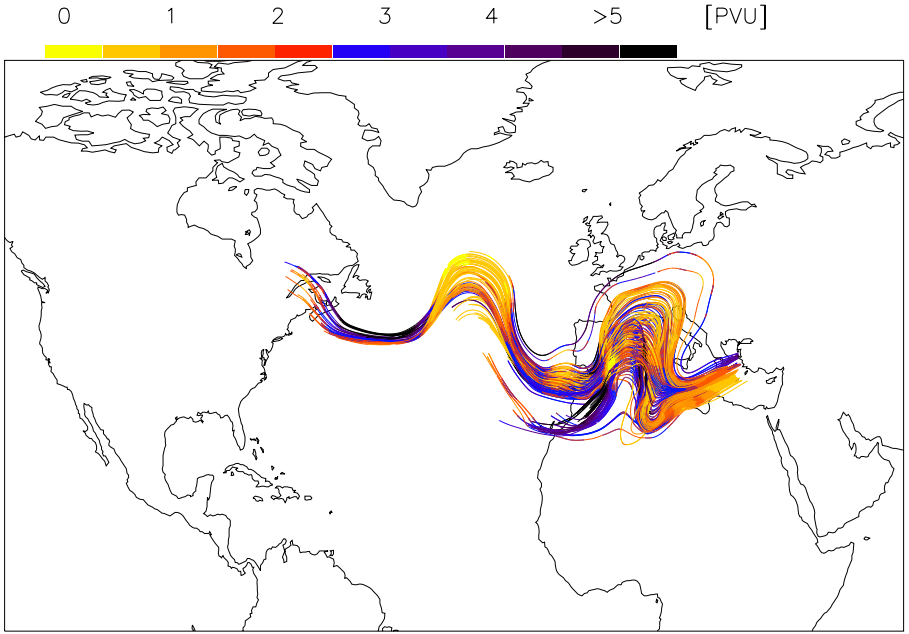

Fig. 4. Potential vorticity of a subset of the North Atlantic Region trajectories. Some air parcels temporarily have potential vorticity values higher than 2 PVU, indicating stratospheric influence.

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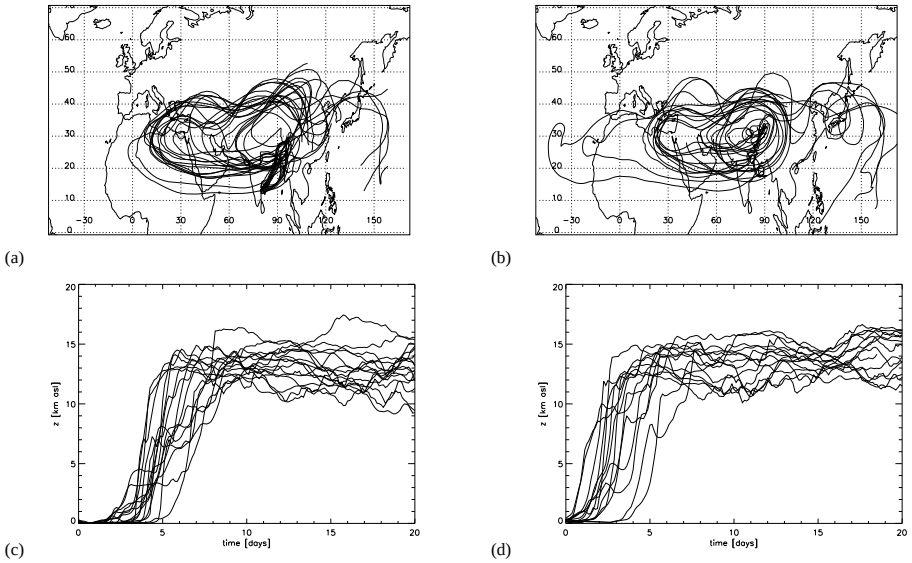

Fig. 5. 20-day forward trajectories for two big Indian cities **(a)** Madras (80.76° E/13.85° N, SE India) and **(b)** Patna (85.72° E/26.01° N, NE India) in July, 2001. Figure 5c shows the temporal development of the trajectory height above sea level for the **(c)** Madras trajectories and **(d)** for those from Patna.