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A case of transatlantic aerosol transport detected at the Schneefernerhaus Observatory (2650 m) on the northern edge of the Alps

WOLFRAM BIRMILI[1]*, TINA GÖBEL[1], ANDRÉ SONNTAG[1], LUDWIG RIES[2], RALF SOHMER[2], STEFAN GILGE[3], INGEBORG LEVIN[4], and ANDREAS STOHL[5]

[1]Leibniz Institute for Tropospheric Research (IfT), Leipzig, Germany
[2]German Federal Environmental Agency (UBA), GAW station Schneefernerhaus, Zugspitze, Germany
[3]German Meteorological Service (DWD), Meteorological Observatory, Hohenpeissenberg, Germany
[4]Institut für Umweltphysik, Universität Heidelberg, Germany
[5]Norwegian Institute for Air Research (NILU), Kjeller, Norway.

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Abstract

An unusual peak in atmospheric aerosol particle number and mass concentration occurred on September 20 and 21, 2006 at the Schneefernerhaus (SFH) Observatory, Germany. A source-receptor analysis using the Lagrangian transport model FLEXPART suggested that polluted air from source regions in the Western and Central U.S. travelled within 4–8 days over the Atlantic to the European Alpine region. A warm conveyor belt over the northern Great Plains was identified as the essential process lifting the boundary layer air to the high altitudes required for a rapid transatlantic transit. The layer arriving at SFH had an aerosol mass concentration of about 30 $\mu g\,m^{-3}$, and a particle number size distribution showing an aged accumulation mode aerosol with mode diameter around 0.3 μm. A combination of in-situ humidity, ^{214}Po and carbon monoxide measurements as well as upper air observations (radiosoundings) suggested that the layer had no previous contact with the local/European boundary layer. A screening of three years of FLEXPART simulations (2005–2007) yielded this case study as the only event of transatlantic anthropogenic aerosol to SFH where mixing with the boundary layer could safely be excluded. The event therefore represents rare surface-based evidence of transatlantic transport of fine and ultrafine aerosols.

Zusammenfassung

Die Verteilung atmosphärischer Spurenstoffe sowie ihre Auswirkungen auf die Luftqualität kann nicht ohne die Betrachtung weitreichender Transportvorgänge verstanden werden. Die Konzentrationen atmosphärischer Aerosolpartikel werden in Bodennähe normalerweise stark durch die Mischungsvorgänge in der Grenzschicht geprägt, weshalb interkontinentaler Ferntransport dort in der Regel nur verwischt wahrgenommen werden kann. Diese Arbeit beschreibt einen Fall bodengestützter Beobachtungen an der Zugspitze (Schneefernerhaus) vom 20. September 2006, bei dem eine Aerosolschicht auf Quellen innerhalb Nordamerikas zurückgeführt werden konnte. Als Transportmechanismus wurde ein warmes Förderband (warm conveyor belt) über den dortigen Great Plains erkannt, welches Grenzschichtluft in die für einen schnellen Transatlantiktransport erforderlichen Höhen brachte. Das ankommende Aerosol war durch ein gealtertes Größenspektrum an Akkumulationsmodepartikeln (Modaldurchmesser 0,3 μm) geprägt und Lagrangeschen Modellrechnungen zufolge zwischen 4 und 8 Tagen aus Nordamerika unterwegs. Betrachtungen der Zerfallsaktivität von ^{214}Po und der Aerosol-, Kohlenmonoxid- und Feuchtemesswerte an der Zugspitze und am tiefergelegenen Hohenpeißenberg sowie Radiosondenprofile zeigten, dass das an der Zugspitze ankommende Aerosol nicht durch Einflüsse aus der lokalen Grenzschicht gestört war. Der Fall verdeutlicht, dass Verschmutzungsaerosol aus Nordamerika in Europa direkt bis an den Boden gelangen kann — wenngleich eine Häufigkeitsanalyse ergab, dass derartiger Direkttransport in die Alpenregion eher selten auftritt (ca. 1/Jahr). Der gezeigte Fall war im Dreijahreszeitraum 2005–2007 der einzige transatlantische Ferntransport, bei dem ein Einfluss der lokalen Grenzschicht auf die gemessenen Partikelkonzentrationen sicher ausgeschlossen werden konnte.

1 Introduction

Atmospheric aerosols are an integral part of the earth's climate system, influencing the earth's radiative balance (HAYWOOD and BOUCHER, 2000; FORSTER et al., 2007), and the oxidation capacity of the atmosphere (RAVISHANKARA, 1997). Ambient aerosols have also been identified as a concern for human health, causing respiratory and cardiovascular disease, particularly in urban areas and megacities (WHO, 2004). Due to the long life-time of fine aerosol particles (diameter < 1 μm) of up to two weeks, natural and anthropogenic aerosols can be transported far downwind of their original source regions. In the beneficial case, long-range transport of mineral dust from arid regions contributes to the fertil-

*Corresponding author: Wolfram Birmili, Leibniz Institute for Tropospheric Research, Permoserstrasse 15, 04318 Leipzig, Germany, e-mail: birmili@tropos.de

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ity of soils in other regions downwind (GRIFFIN et al., 2002). In the less favorable case, such transport may degrade the air quality in normally less polluted regions, or aggravate existing air quality problems. Air quality in North America, for instance, can significantly be influenced by Asian anthropogenic emissions (JAFFE et al., 2003; THORNHILL et al., 2008). In the E.U. and the U.S. the concentrations of airborne particulate matter (PM) are under scrutiny with respect to legal limit values of PM_{10} and $PM_{2.5}$[1] mass concentrations. To devise efficient abatement measures, governments have a vital interest in knowing the relative contributions of local and regional versus remote sources (HOLLOWAY et al., 2003). Consequently, researchers have attempted to apportion surface-measured PM concentrations and/or their exceedances above the legal limit value towards PM source types and regions (HEALD et al., 2006; CHARRON et al., 2007). A general line of arguments is that remote-transported aerosols, even if not discernable specifically, add on top of local and regional pollution (CHIN et al., 2007).

The Central European atmosphere can be influenced by long-range aerosol transport from Africa, North America, and also Asia via North America. Lidar measurements document the frequent transport of Saharan dust (MÜLLER et al., 2003), or forest fire aerosols from regions as far as Canada and Siberia (MÜLLER et al., 2005) into Central Europe. Most long-range transported aerosols have actually been detected in elevated atmospheric layers. Saharan dust, however, can be mixed into the boundary layer north of the Alps and raise PM concentrations at the surface significantly (BRUCKMANN et al., 2008). In apparently rare cases, dust aerosols from distant semi-arid regions like the southern Ukraine may reach Central Europe through the boundary layer (BIRMILI et al., 2008).

Anthropogenic pollution can be transported from North America to Europe through warm conveyor belts (STOHL and TRICKL, 1999). Earlier studies have identified transatlantic transport by aircraft measurements (STOHL et al. 2003, HUNTRIESER et al., 2005). The detection of transport from North America has mostly relied on airborne and/or remote sensing measurements. Evidence from ground-based measurements, in contrast, has been scarce.

In this work, we report on an unusual episode of high concentrations of sub-µm aerosol particles observed at the Schneefernerhaus Observatory in southern Germany in an air mass transported from North America. This observation allowed, for the first time, an in-situ characterization of such a remotely transported aerosol layer.

[1] Particulate matter with aerodynamic diameters smaller than 10 and 2.5 µm, respectively.

2 Methods

2.1 In-situ measurements

The Schneefernerhaus (SFH) observatory (47° 25'N, 10° 59'E, 2656 m a.s.l.) is a WMO-GAW (Global Atmosphere Watch) station located ca. 300 m below the summit of the Zugspitze mountain in southern Germany. SFH hosts a wide range of aerosol and trace gas measurements to characterize air from the boundary layer but also the free troposphere.

Particle number size distributions of ambient aerosols have been measured continuously since December 2004 using a scanning mobility particle sizer (SMPS model 3080, TSI Inc., St Paul, USA). The SMPS measures the size distribution of particles between 10 and 600 nm at a time resolution of 5 min. The SMPS measurements are part of activities within the German Ultrafine Aerosol Network (GUAN), devoted to long-term observations of climate and health-related properties of the environmental aerosol (BIRMILI et al., 2009a). All particle concentrations were normalised to standard temperature (0°C) and pressure (1013 hPa). BIRMILI et al. (2009b) gave a basic overview of the particle size distributions collected at SHF since 2004, exhibiting a median value of around $1000\,\mathrm{cm}^{-3}$ in total particle number concentration, and around $2\,\mu\mathrm{g\,m}^{-3}$ in PM_1 mass concentration. The particle size distributions showed a significant annual cycle with higher concentrations in summer, and lower ones in winter, which was attributed to seasonal changes of the mixed layer height.

Additional measurements at SFH include relative humidity, temperature, wind speed, wind direction, and reactive trace gases. Meteorological parameters were also collected at Zugspitze summit (2964 m a.s.l.), which is in an unobstructed position with respect to all wind directions. Aerosol, gas and meteorological parameters were also collected at the Meteorological Observatory Hohenpeissenberg (HP) – part of the joint WMO-GAW station with SFH but located 40 km to the north at a lower altitude (990 m). Radiosounding ascents were made at Munich, ca. 90 km northeast of the Zugspitze mountain at an altitude of 480 m.

2.2 Lagrangian transport model

The Lagrangian particle dispersion model FLEXPART was used to determine the source regions of air constituents over the Alpine region. FLEXPART is a stochastic particle dispersion model with turbulence and convection parameterizations (STOHL et al., 2005). It was run in backward mode for this study. For the theory underlying the calculations see SEIBERT and FRANK (2004), and for applications to aircraft measurements see STOHL et al. (2003). FLEXPART was driven with model-level analyses with $1^\circ \times 1^\circ$ resolution and 60 vertical layers from the European Centre for Medium-Range Weather Forecasts (ECMWF).

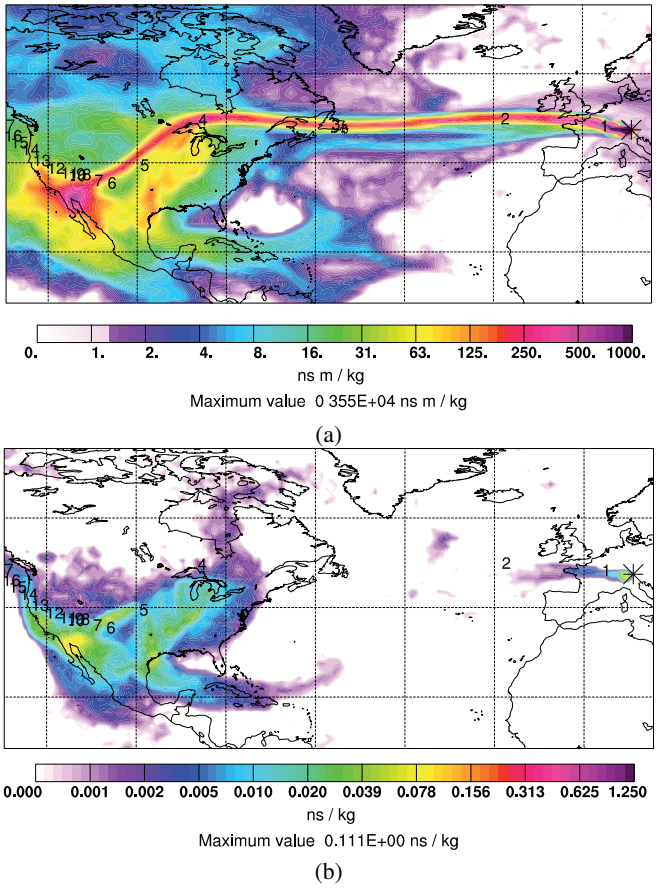

Figure 1: Source-receptor relationship with arrival time at SFH on September 21, 2006, 06:00 UTC: (a) Column-integrated relationship, (b) Footprint relationship. The intensities shown represent gridded emission sensitivities that are proportional to the residence time of air in the respective grid cells.

The model output from the backward simulations consists of gridded emission sensitivities, which are proportional to the residence times of the particles in the grid cells. The emission sensitivity (in units of $\mathrm{s\,kg^{-1}}$) is a measure for the simulated mixing ratio at the receptor that a source of unit strength ($1\,\mathrm{kg\,s^{-1}}$) in the respective grid cell would produce. In this work, emission sensitivities are shown both for the footprint as well as integrated over the entire atmospheric column. The column-integrated emission sensitivity shows the overall pathway of the air towards SFH and can be qualitatively interpreted like simple back trajectories.

The so-called "footprint" refers to the emission sensitivity in the lowest model layer (0–100 m), and is of particular interest because most pollutants are emitted near the surface. Folding (i.e. multiplying) the footprint emission sensitivity with the emission flux densities (in units of $\mathrm{kg\,m^{-2}\,s^{-1}}$) from an appropriate emission inventory yields a so-called source contribution map, which illustrates the geographical distribution of sources contributing to the simulated mixing ratio at SFH under the assumption that chemical species are conserved during transport like a passive tracer. Spatial integration of these source contributions yields the simulated mixing ratio at the receptor.

We use carbon monoxide (CO) as a transport tracer, which is nearly conserved in the atmosphere over the transport times considered here. CO emissions were taken from FROST et al. (2006) for North American emissions and from the EDGAR 3.2 Fast Track 2000 global inventory elsewhere (OLIVIER and BERDOWSKI, 2001). By using a series of backward simulations for SFH every 3 hours, the CO tracer can be displayed as a time series. By integrating the source contributions only over specific areas (e.g., Europe, North America) instead of globally, regional tracer mixing ratios can be obtained. The simulated CO mixing ratios take into account only CO emissions during the period of backward model simulation (20 days) and thus only represent the enhancement over an aged background.

3 Results

3.1 Meteorological transport

3.1.1 Detection of long-range transport

Figure 1 illustrates the source-receptor relationship calculated for arrival times at SFH on September 21, 06:00 UTC. The sub-figure 1a visualizes the zonal transport path from North America to Europe, with a transit time of just 2 days over the Atlantic. Over the ocean, the air mass traveled well above the marine boundary layer, at altitudes between 4 and 6 km before subsiding over the Alpine region. The footprint source-receptor relationship in Fig. 1b, which considers contributions from the model surface layer only, highlights the source regions over North America, where boundary layer air was supposedly mixed into a warm conveyor belt.

Figure 2 illustrates the detection of the event within a monthly context: Simulated concentrations of American as well as European carbon monoxide (CO) tracer (cf. Sect. 2.2). are compared. On Sep 21, 06:00 UTC the simulated American tracer contributions exceeded the European contributions by a factor of 16.6, i.e. the European contributions were predicted to be negligible. Normally the reverse is true; the median ratio between American and European CO tracer contributions for September was 0.037 (roughly 1:27). A closer look at the age spectrum of American CO tracers (not shown) suggested that the majority of CO arriving at SFH was emitted over the U.S. between 6 and 12 days before.

3.1.2 Genesis of the warm conveyor belt

Figure 3 displays the 500 hPa geopotential height on September 17, 00:00 UTC over North America. During the two previous days, an intensive long-wave trough over Western North America expanded in southwesterly direction, and can be seen in Fig. 3 over the U.S. state of Montana. On its eastern fringe, a surface

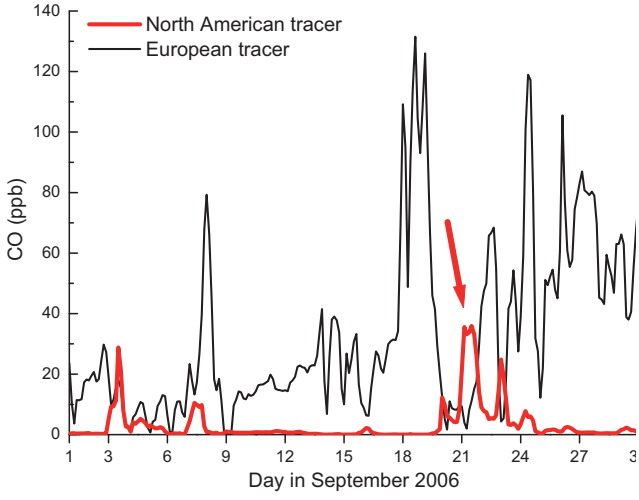

Figure 2: Simulated concentrations of American CO tracer (up t 14 d old) at SFH during September 2006. The peak on Sep 20 21 corresponds to the event of high particle mass concentrations detected at SFH. The concentrations of European CO tracer are plotted for comparison.

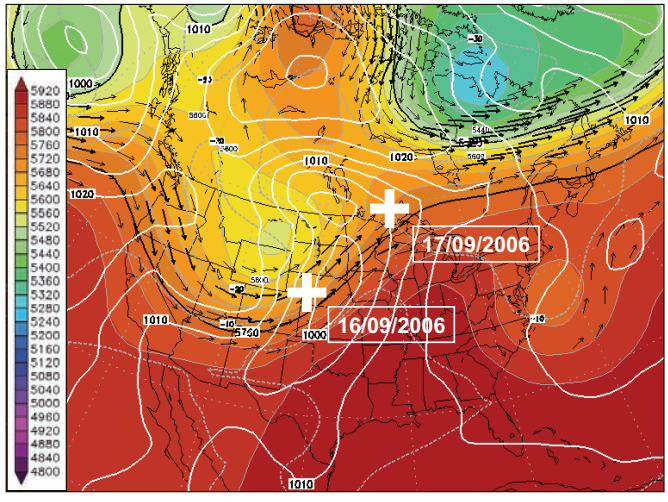

Figure 3: Geopotential height on the 500 hPa level in m over the North American source region, September 17, 2006, 00:00 UTC. The two crosses indicate the centroid location of the air mass sampled at SHF on September 16 and 17, 2007 (00:00 UTC respectively), which was lifted into the conveyor belt. Contour lines indicate sea level pressure in hPa. Source: National Centers for Environmental Prediction (NCEP) reanalysis.

low pressure area was formed over the Great Plains, centered around the state of Nebraska. In this constellation, air from the south-western U.S. was conducted in cyclonic motion around the trough in north-westerly direction. Over the Great Plains, this warm and moist air was forced to slide over the cold air residing in the northern part of the surface low pressure area, thus forming a warm conveyor belt (WCB). According to our Lagrangean model simulation, the lifting of air into the conveyor belt took place between Sep 16 and 17 (00:00 UTC respectively) between the two locations marked by crosses in Fig. 3. As a result, the lifted air reached considerable altitudes between 5 and 8 km on Sep 17 and 18 over the province of Ontario, Canada.

Once at higher altitudes, the air was exposed to the polar front jet stream south of the low pressure system over northeastern Canada. There, the air mass was considerably accelerated and also deflected towards easterly direction. The synoptic constellation was rather favorable for a rapid transatlantic transport because a lengthy high-level trough extended between Canada and Europe between Sep 18 and 19, accounting for a rapid zonal motion (cf. Fig. 1a). Subsequently, the air subsided due to an anticyclone over Central Europe. There, the air mass slowed down considerably, eventually deflecting to southerly directions during the last twelve hours before arrival at Zugspitze.

The lifting process identified in Fig. 3 corresponds to previous descriptions of the genesis of warm conveyor belts (CARLSON, 1980; SCHULTZ, 2001). The FLEXPART simulations of backward transport corresponded well to the North American synoptic situation. Also, the cloud band associated with the WCB could be detected in satellite images (not shown). It is worth to note that the lifting process occurred over the center of the North American continent rather than on its eastern edge, as observed in several previous case studies.

3.1.3 Verification of the air mass origin

Apart from FLEXPART, additional backward and forward trajectories were calculated using the NOAA HYSPLIT model (FNL data set; DRAXLER and HESS, 2004). They yielded essentially the same results and confirmed the southwestern and central U.S. as the source region of the transported aerosols. The centroid location of the plume eventually moving towards Europe was California (Sep 14, 00:00 UTC), the southern Rocky mountain states (Sep 15, 00:00 UTC), the northern Great Plains (Sep 16, 00:00 UTC), and southeastern Ontario (Sep 17, 00:00 UTC). The continental contributions to the air stream conveyed to Europe therefore date from between 4 and 8 days prior to arrival.

It is necessary to note that observations from the satellite-borne Moderate Resolution Imaging Spectroradiometer (MODIS) revealed extensive smoke plumes from wild fires between Sep 15 and 18 over some Northern U.S. states as well as the Canadian provinces Manitoba and Ontario. These fires had prevailed during much of August and September 2006 (SAUNDERS, 2007) but were particularly active during the period of the WCB. It was therefore necessary to consider possible contributions from these biomass burning plumes.

The evaluation of the MODIS images as well as the vertical atmospheric structure gathered from radiosounding ascents suggested that the biomass burning plumes stayed overwhelmingly in the lower troposphere. (In particular, their path followed a more south-easterly

direction compared to the zonal high level jet, with most of the plume blending into the marine boundary layer.) However, due to the significant convection that biomass burning plumes can develop, one cannot exclude that parts of these smoke plumes were mixed into the warm conveyor belt during the last stage of the lifting process over south-western Ontario.

3.2 In-situ observations

The arrival of the polluted aerosol layer was detected at SFH around midnight, between September 20 and 21, 2006. Figure 4 gives an overview of the in-situ measurements during the two days. Aerosol physical, gas phase chemical and meteorological parameters from the three observation sites Schneefernerhaus (SFH, 2650 m), Zugspitze summit ("Zug", 2960 m), and the lower-level Hohenpeissenberg ("HP", 990 m) are included.

The contour diagram of the particle number size distribution shows a multifaceted evolution: In the morning of Sep 20, an accumulation mode with particles mainly between 100 and 300 nm dominates the size spectrum at SFH between 00:00 and 09:00 local winter time (Fig. 4). Between 09:00 and 16:00 an oscillating signal in particle number could be seen, which correlated with nitrogen oxides (not shown). These mid-day peaks occurred in updraft air from southerly wind directions and very likely stemmed from anthropogenic sources (restaurant buildings) near the lower-level plateau "Zugspitzplatt". Between 15:00 and 21:00 a bimodal aerosol distribution can be seen with an accumulation mode (diameter range 100–300 nm) and an Aitken mode (range 10–40 nm).

Around 22:00 the size distribution changes significantly, leaving an accumulation mode of increased mean diameter as well as of enhanced concentration. This air mass, which prevails until about 06:00 on the next day, is what we identified as the polluted aerosol layer from North America. In Fig. 4 this period is marked by arrows.

Integral particle number concentrations for three different size intervals suggest the transport event to be essentially connected with larger particles: While the number of Aitken particles (30–60 nm) decreased, the number of accumulation mode particles (150–600 nm) increased significantly. From the particle number size distribution, a PM_1 particle mass was estimated assuming a material density of $1.6\,g\,cm^{-3}$. The time series of calculated PM_1 confirms the remarkable peak at Sep 20, 22:00, which reaches a maximum value of $36\,(\pm 5)\,\mu g\,cm^{-3}$.

Importantly, the time series of PM_{10} particle mass recorded at the lower HP site shows no such peak at all. This strongly suggests that the high particle mass concentrations came from aloft rather than from the local boundary layer. The observations of particle mass are supported by those of carbon monoxide (CO): During the night of Sep 20/21, CO values reached levels between 200 and 250 ppm at SFH, whereas the HP concentrations stayed around 130 ppm throughout the night. The comparison between the two sites allows to define the transport event to the period Sep 20, 20:00 – Sep 21, 06:00. The polluted character of the air mass in the elevated layer was confirmed by slightly increased levels of ozone (O_3) and elevated levels of peroxyacetyl nitrate (PAN; 0.6 ppb compared to a background level of 0.3 ppb).

An instrumental observation was the decay rate of the radionuclide ^{214}Po, a short-lived daughter product of ^{222}Rn. In equilibrium, the decay rate of ^{214}Po is proportional to the concentration of radon, which in turn is a tracer for near-surface air (STOCKWELL and CHIPPERFIELD, 1999; LEVIN et al., 2002). The half-life time of radon is 3.8 days, its main source being rocks and soil. Its suitability as a tracer of near-surface air derives from its overwhelming independence of anthropogenic processes.

At the time of the transport event, the decay rate of ^{214}Po decreased rapidly, from around 2 down to about $0.1\,Bq\,m^{-3}$ (Fig. 4). Typical levels of the decay rate are $3\,Bq\,m^{-3}$ in summer and $0.5\,Bq\,m^{-3}$ in winter (BIRMILI et al., 2009b). This drop in the ^{214}Po decay rate is in sharp contrast to the observed increase in particle mass concentration, CO and PAN – all indicators of pollution. The lack of radon is an unusual feature in the whole body of observations and can only be explained by the previous detachment of the arriving air from the surface. For completeness, we add that the relative humidity at SFH was only 27 %, i.e. there was no possibility for the presence of cloud droplets. This ensures that there were no loss terms for ^{214}Po except radioactive decay, i.e. the ^{214}Po measurement is proportional to the concentration of radon. The decrease in radon in conjunction with increasing CO is the most significant evidence for the origin of the observed air in North America.

A further indicator for remote transport is the decline in specific humidity s from around $4\,g\,kg^{-1}$ down to $2\,g\,kg^{-1}$ after Sep 20, 20:00. The local meteorological parameters were rather constant during the entire night, suggesting a coupling of the SFH site to the geostrophic winds. The dry temperature was steadily around 6.5° C. The wind speed was constantly around $6\,m\,s^{-1}$ at the Zugspitze summit, and somewhat lower at SFH ($4\,m\,s^{-1}$). The winds were consistently blowing from westerly direction both, at SFH and the summit. The vertical wind velocity at SHF (mean value $0.5\,m\,s^{-1}$) indicates a continuous subsidence of the arriving air, which is in marked contrast to the usual thermal updrafts during daytime due to SFH's location on the southern slope of the Zugspitze mountain (GANTNER et al., 2003).

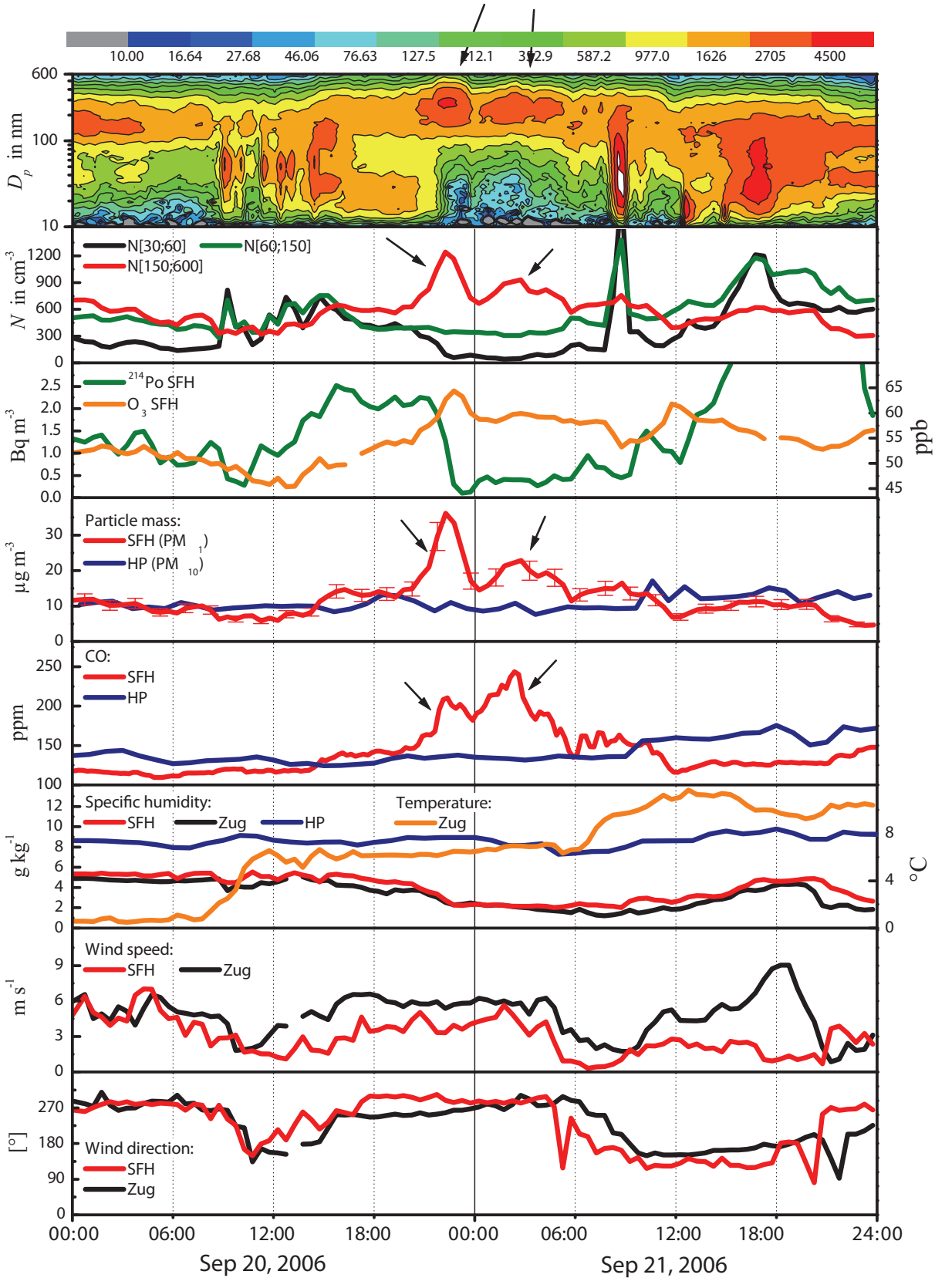

Figure 4: In situ observations on September 20 and 21, 2006 at Schneefernerhaus ("SFH", 2650 m a.s.l.), the Zugspitze summit ("Zug", 2960 m), and Hohenpeissenberg ("HP", 990 m): Particle number size distributions at SFH in dN/dlogDp and cm^{-3}; particle number concentration integrated over diameter intervals 30–60, 60–150, and 150–600 nm at SFH; decay rate of ^{214}Po and ozone (O_3) mixing ratio at SFH; PM mass concentrations at HP (directly measured) and SFH (calculated); mixing ratio of carbon monoxide (CO) at SFH and HP; specific humidity at all sites and temperature at Zug; wind speed and wind direction at SFH and Zug. Times are indicated in local time (CET). All concentrations refer to standard temperature (0°C) and pressure (1013 hPa). Presumed transport contributions from North America are marked by arrows.

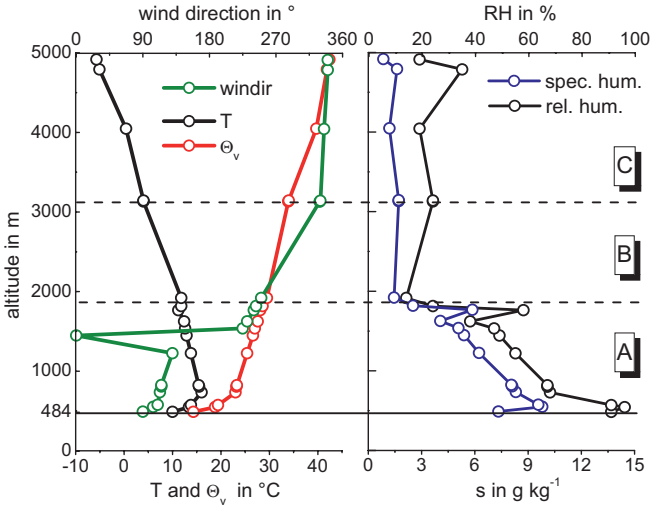

Figure 5: Radiosounding ascent at Munich on Sep 21, 2006, 00:00 UTC. Three distinct layers can be seen below 4000 m. Layer 2 = B, between an altitude of 2000 and 3000 m corresponds to the aerosol layer detected at SFH. Layer 1 = A is the residual layer below 2000 m altitude, which has a depth of 1500 m. Layer 3 = C originates from north-westerly directions and has not been identified yet.

The concentration of nitrogen monoxide (NO, not shown) was below the detection limit (0.025 ppb) throughout the night, which confirms that there was no fresh emission input into the sampled air mass. The concentration of total reactive odd nitrogen (NO_y), however, was enhanced, ranging between 0.7-1.0 ppb. This indicates that nitrogen oxides were nearly completely oxidized to higher oxidized NO_y species, again pointing towards a polluted but aged air mass that received no fresh emission input. The NO_y concentrations in the North American plume were clearly enhanced over a clean free tropospheric background; in the European boundary layer, the values can be significantly higher.

In Sec. 3.1.3 we raised the question of contributions from anthropogenic sources in the southwestern U.S. versus those from the evident wild fires in the Canadian provinces of Manitoba and Ontario. From a closer look at the ratios of CO, sulphur dioxide (SO_2) and sulphur hexafluoride (SF_6) one would expect to gain information on the sources of the aerosols. As an essential finding, the ratios [SO_2]/[CO] and [SF_6]/[CO] appeared to decrease at SFH by roughly 50 % during the transport event compared to European-influenced air. As both SO_2 and SF_6 are primarily of anthropogenic origin, it seems likely that biomass burning aerosols contributed to the transatlantic plume. This would, in fact, not be surprising given the frequent occurrence of North American wild fires particularly in 2006 (SAUNDERS, 2007).

3.3 Vertical structure over the Alpine region

While the trajectory calculations confirmed a clear upward motion over North America and then subsidence over Europe, we employed vertical information from radiosoundings to confirm the multi-layered structure of the atmosphere over the Alps during the transport event. Figure 5 illustrates the radiosounding data recorded at Munich on Sep 21, 00:00 UTC (90 km northeast of the Zugspitze mountain and located at an altitude of about 480 m). Three distinct layers can be seen below 4000 m: Layer B, between an altitude of 2000 and 3000 m corresponds to the aerosol layer detected at SFH. Layer A is the residual layer below 2000 m altitude, which has a depth of 1500 m. A vertical gradient can be seen in specific humidity s: 1.7 g kg^{-1} above an inversion located at an altitude of 2000 m but more humid air in the boundary layer below (5–10 g kg^{-1}). Layer C originates from rather northerly than westerly directions and has not been studied any further. Between layer A and B an enhanced temperature inversion can be seen. The verification of the radiosounding ascent at Innsbruck (south of SFH) on Sep 21, 03:00 UTC confirmed the same vertical structure. The radiosounding observations add to the impression that the remotely transported aerosol layer was wholly detached from the boundary layer and thus arrived at SFH rather uninfluenced from European sources.

3.4 Aerosol characteristics in the remotely transported layer

Figure 6 illustrates the particle number size distribution at SFH during the transport event. The observed size distribution indicates a rather aged aerosol, i.e. being practically devoid of small particles, and with a maximum in the number concentration close to 300 nm. A least-squares fit procedure (BIRMILI ET AL., 2001) was applied to describe the number size distribution as a composite of multiple lognormal modes. This procedure yielded, as an average for the distribution at SFH between 22:00 and 06:00, the following lognormal modal parameters: accumulation mode I (geometric mean diameter: 288 ($\pm$ 8) nm; number concentration N: 2600 ($\pm$ 240) cm^{-3}; spread parameter: 1.41), accumulation mode II (153 ($\pm$ 13) nm; 1030 ($\pm$ 200) cm^{-3}; 1.29), Aitken mode (79 ($\pm$ 9) nm; 440 ($\pm$ 100) cm^{-3}; 1.49).

It is worth to note that the accumulation mode diameter of 288 nm belongs to the upper range of accumulation mode observations: In the Central European boundary layer, the most frequent accumulation mode diameter range is 190-210 nm (BIRMILI et al., 2001). Bigger modal diameters ($>$ 280 nm) have been identified to represent rather aged aerosols (BIRMILI et al., 2001), also including a droplet mode that most likely results from cloud processing within the continental boundary layer (COSTABILE et al., 2009). This result adds to the impression that the observed aerosol is an aged anthropogenic plume.

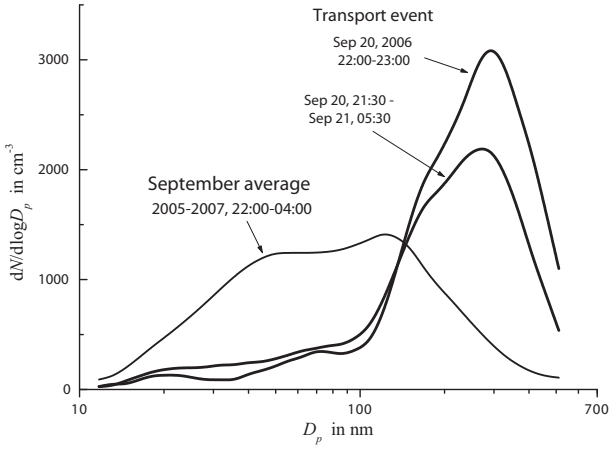

Figure 6: Particle number size distributions at SFH during the transport event, in comparison with the background values typical for September nights at SFH.

4 Discussion

There is climatological evidence that the transport of atmospheric pollution from North America to Europe takes place mainly through warm conveyor belts (WCBs) (STOHL, 2001; STOHL et al., 2002). Continental air over North America may be lifted by convection, and travel through the middle and upper troposphere in the form of mid-level and high-level jet streams (HUNTRIESER et al., 2005).

WCBs are associated with frontal systems and belong to the most relevant mechanisms for the transport of warm air masses in general (COOPER ET AL., 2004). WCBs are located on the warm side of a jet stream and, thus, also responsible for the clouds and precipitation ahead of the surface cold front. WERNLI AND DAVIES (1997) pointed out the strong bundling of trajectories in association with warm conveyor belts, and the limited mixing that occurs between the transported air mass and the surrounding air.

From a global climatological point of view, WCB air parcels originate most frequently in the boundary layer over the warm water pools at the eastern seaboards of North America and Asia (STOHL, 2001). As a result of the substantial population and industrial centres located there, the conveyor belts play an important role in the global dispersion of pollution. Because WCBs rarely start over Europe, European pollution has fewer chances of reaching the upper troposphere and becoming globally distributed compared to the emissions from other densely populated regions. Over the past few years, the relevance of WCBs has been confirmed in many case studies and climatological studies (cf. above references). Cases of pollution export from North America have been observed at low altitudes (e.g., behind a cold front) but those plumes normally do not reach Europe as a coherent and therefore rather undiluted plume.

One could also think of transatlantic transport without WCBs, e.g., in a warm, dry and polluted air mass moving adiabatically from the southwestern U.S. into the middle troposphere where the air mass is then rapidly transported towards Europe. Although this process may in principle be possible, it seems to occur only rarely. We are not aware of a single publication demonstrating this pathway. It appears that such an uplift is not strong enough to subject a polluted air mass to a fast enough air stream, for the air mass to be transported coherently to Europe.

The case of transatlantic transport presented in this paper shows all relevant features of a WCB described before, and can thus be considered a relevant case for transatlantic pollution transport. Climatologies show that WCBs ascend most frequently over the North Atlantic downwind of the North American east coast. However, in summer, while generally less frequent than in winter, they often also originate over the North American continent itself (ECKHARDT et al., 2004). The case presented here is an example for such a WCB associated with a cyclone over North America. A previously unseen feature is that the elevated aerosol layer could be characterized in detail at a ground-based observation site in Europe.

To scrutinize on the overall relevance of such transatlantic transport events, Lagrangian transport simulations for the entire period between 2005 and 2007 were examined. However, only a surprisingly small number of clear transport events from North America to Europe could be identified. Based on the prerequisite of a substantial footprint of the back trajectory over the North American continent, only two other transport events were determined: April 28/29, 2005, and August 25, 2005. A detailed look at those case studies revealed, however, a confounding influence of European boundary layer air. This judgement was made on the basis of the substantial ^{214}Po decay rates, which would be expected near zero in case of a transatlantic transport event. The event presented in this paper represents the most pronounced and most clearly detectable transport event over these three years on the basis of the SFH in situ data.

A last question concerns whether wet scavenging might have had an effect on the aerosol observed. Since the uplift occurred over the U.S. in a warm conveyor belt, one needs to take into consideration nucleation scavenging, i.e. cloud activation of particles and their subsequent removal by rain-out. Surface weather observations reported light rain in the northern section of the region where the uplift occurred (U.S. states of South and North Dakota, and Minnesota), although not upstream of these areas. The particle size distribution in Fig. 6 denies the effectiveness of nucleation scavenging, which would cause a depletion of particles especially in the accumulation mode (> 100 nm). Two explanations seem possible: Either, nucleation scavenging in the ascending air parcel was not effective enough, for instance,

because the hydrometeors were still too small to cause sufficient precipitation; or, the entrainment of biomass burning particles over Canada in the later stage of the plume was the predominant feeding process for surface-emitted aerosol. Within the scope of the available data we feel not able to make a final conclusion here.

5 Conclusions

An unusual peak in atmospheric aerosol particle number and mass concentration was detected on September 20 and 21, 2006 at the Schneefernerhaus Observatory in the German Alps, and could be tracked to source regions in North America. A warm conveyor belt over the northern Great Plains was identified as the essential lifting mechanism for a warm sector air mass from the North American boundary layer that was subsequently transported rapidly across the Atlantic in a zonal high-level jet. The south-western and Central U.S. could be identified as a prime source region of the aerosol, while additional contributions by wild fires in the Canadian provinces of Manitoba and Ontario – mixed into the lifted stream by convection, appear likely based on an analysis of the ratios of sulphur dioxide and sulphur hexafluoride compared to carbon monoxide. Although the aerosol mass concentration in the plume arriving in Europe was moderate ($30\,\mu g\,m^{-3}$) compared to the legal limit value of PM_{10} (daily averages $50\,\mu g\,m^{-3}$), the observation highlights the possibility of American continental aerosol to reach European ground in a rather undiluted fashion. The observed particle size distributions showed an accumulation mode with mean diameter around 300 nm, which is a consistent indicator of an aged pollution aerosol as well as an aged biomass burning aerosol. Combined evidence from in-situ CO, humidity and ^{214}Po measurements, concurrent observations at the lower-level Hohenpeissenberg site, as well as radiosounding observations suggested that the polluted layer was not in contact with the European and/or local boundary layer. This case represents, to our knowledge, one of the first surface-detected cases of transatlantic transport of fine and ultrafine aerosol particles. A screening of three years of FLEXPART simulations (2005–2007), however, yielded this event as the only case when North American pollution aerosol reached the Alpine region in an undiluted fashion. Open questions remain about the role of wet scavenging in limiting the remote-transported aerosol fraction as well as the climatological importance of intercontinental transport paths of air pollution in the northern hemisphere.

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References

BIRMILI, W., A. WIEDENSOHLER, J. HEINTZENBERG, K. LEHMANN, 2001: Atmospheric particle number size distribution in Central Europe: Statistical relations to air masses and meteorology. — J. Geophys. Res. **D23**, 32 005-32 018.

BIRMILI, W., K. SCHEPANSKI, A. ANSMANN, G. SPINDLER, I. TEGEN, and 12 others, 2008: A case of extreme particulate matter concentrations over Central Europe caused by dust emitted over the southern Ukraine. – Atmos. Chem. Phys. **8**, 997–1016.

BIRMILI, W., K. WEINHOLD, S. NORDMANN, A. WIEDENSOHLER, G. SPINDLER, and 14 others, 2009a: Atmospheric aerosol measurements in the German Ultrafine Aerosol Network (GUAN): Part 1 – soot and particle number size distributions. – Gefahrst. Reinh. Luft **69**, 137–145.

BIRMILI, W., L. RIES, R. SOHMER, A. ANASTOU, A. SONNTAG, K. KÖNIG, I. LEVIN, 2009b: Fine and ultrafine aerosol particles at the GAW station Schneefernerhaus/Zugspitze. – Gefahrst. Reinh. Luft **69**, 31–35.

BRUCKMANN, P., W. BIRMILI, W. STRAUB, M. PITZ, D. GLADTKE, U. PFEFFER, H. HEBBINGHAUS, S. WURZLER, A. OLSCHEWSKI, 2008: An outbreak of Saharan dust causing high PM10 levels north of the Alps. – Gefahrst. Reinh. Luft **68**, 490–498.

CARLSON, T.N., 1980: Air flow through mid-latitude cyclones and the comma cloud pattern. – Mon. Weather Rev. **108**, 1498–1509.

CHARRON, A., R.M. HARRISON, P. QUINCEY, 2007: What are the sources and conditions responsible for exceedences of the 24 h PM_{10} limit value (50 µg m^{-3}) at a heavily trafficked London site? – Atmos. Environ. **41**, 1960–1975.

CHIN, M., T. DIEHL, P. GINOUX, W. MALM,, 2007: Intercontinental transport of pollution and dust aerosols: implications for regional air quality. – Atmos. Chem. Phys.**7**, 5501–5517.

COOPER, O. R., C. FORSTER, D. PARRISH, M. TRAINER, E. DUNLEA, and 10 others, 2004: A case study of transpacific warm conveyor belt transport: Influence of merging airstreams on trace gas import to North America. – J. Geophys. Res. **109**, D23S08, DOI:10.1029/2003JD003624.

COSTABILE, F., W. BIRMILI, S. KLOSE, T. TUCH, B. WEHNER, A. WIEDENSOHLER, U. FRANCK, K. KÖNIG, A. SONNTAG, 2009: Spatio-temporal variability and principal components of the particle number size distribution in an urban atmosphere. – Atmos. Chem. Phys. **9**, 3163–3195.

DRAXLER, R., G. HESS, 2004: Description of the HYSPLIT4 modeling system, NOAA Technical Memorandum, ERL, ARL-224.

ECKHARDT, S., A. STOHL, H. WERNLI, P. JAMES, C. FORSTER, N. SPICHTINGER, 2004: A 15-year climatology of warm conveyor belts. – J. Climate **17**, 218–237.

FORSTER, P., V. RAMASWAMY, P. ARTAXO, T. BERNTSEN,, R. BETTS et al., 2007: Changes in Atmospheric Constituents and in Radiative Forcing. – In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. SOLOMON, S., D. QIN, M. MANNING, Z. CHEN, M. MARQUIS, K.B. AVERYT, M. TIGNOR, H.L. MILLER (Eds.), Cambridge University Press, Cambridge (UK), available at www.ipcc.ch

FROST, G.J., S.A. MCKEEN, M. TRAINER, T.B. RYERSON, J.A. NEUMAN et al., 2006: Effects of changing power plant NO_x emissions on ozone in the eastern United States: Proof of concept. – J. Geophys Res. **111**, D12306, DOI:10.1029/2005JD006354.

GANTNER, L., M. HORNSTEINER, J. EGGER, G. HARTJENSTEIN, 2003: The diurnal circulation of Zugspitzplatt: observations and modelling – Meteorol. Z. **12**, 95–102.

GRIFFIN, D.W., C.A. KELLOGG, V.H. GARRISON, E.A. SHINN, 2002: The Global Transport of Dust – American Scientist **90**, 228–235.

HAYWOOD, J., O. BOUCHER, 2000: Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review. – Rev. Geophys. **38**, 513–543.

HEALD, C.L., D.J. JACOB, R.J. PARK, B. ALEXANDER, T.D. FAIRLIE, R.M. YANTOSCA, D.A. CHU, 2006: Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States. – J. Geophys. Res. **111**, D14310, DOI:10.1029/2005JD006847.

HOLLOWAY, T., A. FIORE, M.G. HASTINGS, 2003: Intercontinental transport of air pollution: Will emerging science lead to a new hemispheric treaty? – Env. Sci. Technol. **37**, 4535–4542.

HUNTRIESER, H., J. HELAND, H. SCHLAGER, C. FORSTER, A. STOHL, H. AUFMHOFF, F. ARNOLD, H. E. SCHEEL, M. CAMPANA, S. GILGE, R. EIXMANN, O. COOPER, 2005: Intercontinental air pollution transport from North America to Europe: Experimental evidence from aircraft measurements and surface observations. – J. Geophys. Res. **110**, DO1305, DOI:10.1029/2004JD005045.

JAFFE, D., I. MCKENDRY, T. ANDERSON, H. PRICE, 2003: Six "new" episodes of trans-Pacific transport of air pollutants. – Atmos. Env. **37**, 391–404.

LEVIN, I., M. BORN, M. CUNTZ, U. LANGENDÖRFER, S. MANTSCH, T. NAEGLER, M. SCHMIDT, A. VARLAGIN, S. VERCLAS, D. WAGENBACH, 2002: Observations of atmospheric variability and soil exhalation rate of radon-222 at a Russian forest site: Technical approach and deployment for boundary layer studies. – Tellus **54B**, 462–475.

MÜLLER, D., I. MATTIS, U. WANDINGER, A. ANSMANN, D. ALTHAUSEN, O. DUBOVIK, S. ECKHARDT, A. STOHL, 2003. Saharan dust over a Central European EARLINET-AERONET site: Combined observations with Raman lidar and Sun photometer. – J. Geophys. Res. **108**, 4345, DOI: 10.1029/2002JD002918.

MÜLLER, D., I. MATTIS, U. WANDINGER, A. ANSMANN, D. ALTHAUSEN, A. STOHL, 2005. Raman lidar observations of aged Siberian and Canadian forest fire smoke in the free troposphere over Germany in 2003: Microphysical particle characterization. – J. Geophys. Res. **110**, D17201, DOI:10.1029/2004JD005756.

OLIVIER, J.G.J., J.J.M. BERDOWSKI, 2001. Global emissions sources and sinks. – In: BERDOWSKI, J., R. GUICHERIT, B.J. HEIJ (Eds.): The Climate System, 33–78, A.A. Balkema Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands.

RAVISHANKARA, A.R., 1997: Heterogeneous and multiphase chemistry in the troposphere. – Science **5315**, 1058–1064.

SAUNDERS, G., 2007: How weather helped shape Ontario's 2006 fire season – Weatherwise **60**, 18–24.

SCHULTZ, D.M., 2001: Reexamining the cold conveyor belt – Mon. Wea. Rev. **129**, 2205-2225.

SEIBERT, P., A. FRANK, 2004: Source-receptor matrix calculation with a Lagrangian particle dispersion model in backward mode. – Atmos. Chem. Phys. **4**, 51–63.

STOCKWELL, Z.D., M.P. CHIPPERFIELD, 1999: A tropospheric chemical transport model: Development and validation of the model transport schemes. – Quart. J. Roy. Meteor. Soc. **125**, 1747–1783.

STOHL, A., 2001. A one-year Lagrangian "climatology" of airstreams in the northern hemisphere troposphere and lowermost stratosphere. – J. Geophys. Res. **106**, 7263–7279.

STOHL, A., T. TRICKL, 1999: A textbook example of long range transport: Simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe. – J. Geophys. Res. **104**, 30445–30462.

STOHL, A., S. ECKHARDT, C. FORSTER, P. JAMES, N. SPICHTINGER, 2002: On the pathways and timescales of intercontinental air pollution transport. – J. Geophys. Res. **107**, 4684, DOI:10.1029/2001JD001396.

STOHL, A., C. FORSTER, S. ECKHARDT, N. SPICHTINGER, H. HUNTRIESER, J. HELAND, H. SCHLAGER, S. WILHELM, F. ARNOLD, O. COOPER, 2003. A backward modeling study of intercontinental pollution transport using aircraft measurements. – J. Geophys. Res. **108**, 4370, DOI:10.1029/2002JD002862.

STOHL, A., C. FORSTER, A. FRANK, P. SEIBERT, G. WOTAWA, 2005. Technical Note: The Lagrangian particle dispersion model FLEXPART version 6.2. – Atmos. Chem. Phys. **5**, 2461–2474.

THORNHILL, K. L., G. CHEN, J. DIBB, C. E. JORDAN, A. OMAR, and 10 others, 2008: The impact of local sources and long-range transport on aerosol properties over the northeast U.S. region during INTEX-NA, – J. Geophys. Res. **113**, D08201, Doi:10.1029/2007JD008666.

WERNLI, H., H.C. DAVIES, 1997. A Lagrangian-based analysis of extratropical cyclones, I - The method and some applications. – Quart. J. Roy. Meteor. Soc. **123**, 467–489.

WHO, 2004. Health effects of air pollution. Results from the WHO Project "Systematic review of health aspects of air pollution in Europe". – WHO Report E83080, June 2004, Geneva.