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European surface ozone in the extreme summer 2003

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[1] Measurements of ozone and other trace species in the European EMEP network in 2003 are presented. The European summer of 2003 was exceptionally warm, and the surface ozone data for central Europe show the highest values since the end of the 1980s. The 95th percentiles of daily maximum hourly ozone concentrations in 2003 were higher than the corresponding parameter measured in any previous year at many sites in France, Germany, Switzerland and Austria. In this paper we argue that a number of positive feedbacks between the weather conditions and ozone contributed to the elevated surface ozone. First, we calculated an extended residence time of air parcels in the atmospheric boundary layer for several sites in central Europe. Second, we show that it is likely that extensive forest fires on the Iberian Peninsula, resulting from the drought and heat, contributed to the peak ozone values in north Europe in August. Third, regional-scale model calculations indicate that enhanced levels of biogenic isoprene could have contributed up to 20% of the peak ozone concentrations. Measurements indicate elevated concentrations of isoprene compared to previous years. Sensitivity runs with a global chemical transport model showed that a reduction in the surface dry deposition due to drought and the elevated air temperature both could have contributed significantly to the enhanced ozone concentrations. Because of climate change, such heat waves may occur more frequently in the future and may gradually overshadow the effect of reduced emissions from anthropogenic sources of VOC and NO_x in controlling surface ozone.

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1. Introduction

[2] The European summer of 2003 was exceptionally warm, in particular in central Europe. On the basis of a temperature reconstruction of monthly (back to 1659) and seasonal (from 1500 to 1658) temperature fields for European land areas (25°W to 40°E, 35°N to 76°N) *Luterbacher et al.* [2004] concluded that the summer of 2003 was very likely warmer than any other summer back to 1500. Compared to the 1901–1995 average surface temperature, the summer of 2003 exceeded that average by about 2°C.

[3] The observations reported within EEA (European Environment Agency) show exceptionally long-lasting and spatially extensive episodes of high ozone concentrations, mainly in the first half of August, and covering the regions with the highest temperatures [*Fiala et al.*, 2003]. For all monitoring sites the average number of hourly ozone exceedances above 180 μg m^{-3} was higher in the summer of 2003 than in any of the previous 12 years. Rural monitoring data from the EMEP (European Monitoring and Evaluation Program) network [*Solberg et al.*, 2005] indicate that in central Europe an indicator like the 6-months AOT40 (Accumulated Ozone exposure over a Threshold of 40 ppb) was higher in 2003 than in any other year since 1990, and that the AOT40 values in 2003 were almost a factor 2 higher than the average during the 1990s. In Switzerland the 2003 summer mean of the daily ozone maxima exceeded the 1992–2002 summer mean of daily ozone maxima by more than 15 ppb, corresponding to 5 standard deviations of the 1992–2002 summer means, similar to the deviation in surface temperature [*Ordonez et al.*, 2005]. In the UK the heat wave was studied through the TORCH campaign [*Lee et al.*, 2006], one of the most highly instrumented to date, local ozone formation rates up to 17 ppb h^{-1} was found.

[4] The high levels of atmospheric pollutants had direct consequences for human health. In case studies of the number of deaths related to the 2003 heat wave in the United Kingdom and the Netherlands, *Stedman* [2003] estimated that for the first two weeks of August 2003 there were 2045 excess deaths over the 1998–2002 average, and between 423 and 769 of these were related to elevated ambient ozone and PM_{10} concentrations. In a similar study for the Netherlands, *Fischer et al.* [2003] found an excess of 1000–1400 deaths during the summer of 2003, and 400–

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Figure 1. Monitoring sites used for calculating air mass back trajectories. The region marks the domain defining the European residence times.

600 were air pollution related (ozone and PM_{10}). *Trigo et al.* [2005] have shown that in France the geographical pattern of the temperature anomaly matched particularly well the mortality rates.

[5] *Schär et al.* [2004] and *Beniston* [2004] carried out regional climate simulation calculations to investigate if summers like the one in 2003 could become more prevalent under future climate change. On the basis of the SRES A2 transient greenhouse gas scenario as specified by IPCC, *Schär et al.* [2004] found that for 2071–2100 conditions (SCEN) for a grid point in northern Switzerland for June–July–August (JJA) the JJA average is shifted 4.6°C toward warmer temperatures, and there is a pronounced widening of its statistical distribution with the standard deviation increasing by 102%. This widening is highly significant. Furthermore, they analyzed other models and greenhouse gas scenarios and found that all exhibit a substantial increased level of variability over large parts of Europe. *Schär et al.* [2004] conclude that in response to greenhouse gas forcing the year-to-year variability in European summer climate may increase, and that the unusual European summer of 2003 may be an example of what is to come. *Beniston* [2004, p. 1] draws a similar conclusion: "For many purposes the 2003 event can be used as an analogue of future summers in coming decades in climate impacts and policy studies."

[6] In the following study we present surface ozone data from the EMEP network of rural background sites in Europe, as well as complementary measurements of isoprene and a number of ozone soundings. Ozone in the atmospheric boundary layer is controlled by the emissions of NO_x and VOC and their photochemical transformations, which are temperature-dependent, the residence time of air parcels in the boundary layer over the source regions, the dry removal to the ground and the incoming UV and shortwave visible radiation as well as the horizontal and vertical advection and mixing.

[7] The link between climate change and regional air quality is a topic receiving increased attention the last years. It was a basis for, e.g., the ICARTT campaign recently reported [*Fehsenfeld et al.*, 2006] and is the underlying focus of the study presented below. We propose that a number of positive feedback mechanisms linked to the heat waves contributed to the high surface ozone concentrations observed, and investigate their potential strengths through chemical transport model (CTM) calculations.

2. Meteorological Conditions

[8] The meteorological conditions during the European summer 2003 have been extensively described in several recent studies, and we will only highlight some key points. Over Europe, persistent anticyclonic anomalies, high temperatures and a series of intense heat waves characterized the summer [*Schär et al.*, 2004; *Stott et al.*, 2004]. In particular, several pronounced heat waves led to soaring ground temperatures in August. The hottest period was around the middle of August, during a highly anomalous blocking event centered over France [*Grazzini et al.*, 2003; *Black et al.*, 2004; *Fink et al.*, 2004]. All-time temperature records then tumbled over much of Europe. *Schär et al.* [2004] showed that the temperatures in the anticyclone over central Europe were four standard deviations above normal.

[9] During the heat waves in June and August monthly temperature anomalies of 6–7°C compared to the 1961–1990 climatological average were observed at sites in Switzerland and south Germany. At Hohenpeißenberg, Germany, the period June–August was 5°C higher than the average since 1781 when temperature monitoring began. According to *Fink et al.* [2004] the elevated temperatures in August were not accompanied by increased convection and vertical mixing. On the contrary the August heat wave lead to a stabilization and subsidence, presumably because of the soil drought leading to low humidity in the lower troposphere. The central Europe anticyclonic anomaly was part of an extensive, quasi-stationary Rossby wave train, stretching from the western Atlantic across Europe and toward Siberia. These waves extended high into the stratosphere where they disturbed the easterly circumpolar circulation up to an altitude of 20–25 km [*Orsolini and Nikulin*, 2006].

[10] In addition, the extreme surface temperatures gave the heat wave devastating impact. These high temperatures resulted from a local radiative budget influenced by the lasting spring-to-summer dryness, by the low background soil moisture, and the clear skies during the anticyclonic conditions [*Schär et al.*, 2004; *Black et al.*, 2004]. Another point worth mentioning is that the Atlantic jet stream was displaced northward, resulting in a reduction in the passage of cyclones over central Europe.

[11] On the basis of the FLEXTRA three-dimensional air mass back trajectories [*Stohl et al.*, 1995; *Stohl and Seibert*, 1998] we have estimated the residence times in the European planetary boundary layer (PBL) for air masses arriving at a number of monitoring sites in Europe as shown in Figure 1. This was done by computing the number of hours the trajectories stayed within a central European domain and below a vertical level which was set to 2.5 km above sea level. The domain was defined to be the area between 10°E, 30°W, 35°N and 55°N (Figure 1). The basis for the calculations were 7-d backward trajectories arriving every 6 h at 500 m above ground for the period 1996–2003 for the given sites.

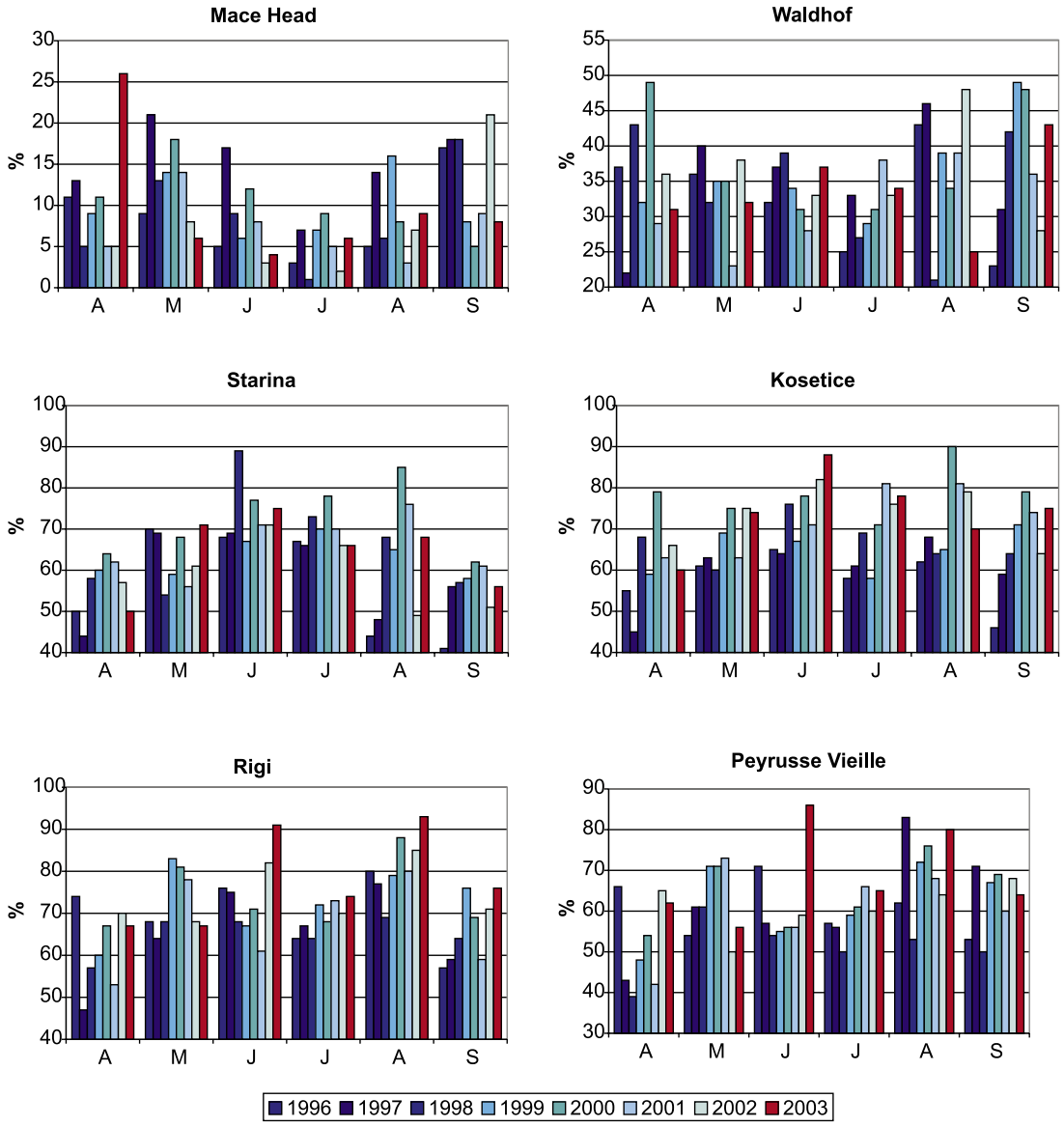

Figure 2. The percentage residence time inside the European planetary boundary for air masses arriving at six sites given as monthly averages (April–September) during 1996–2003. The data are based on 7 d FLEXTRA 3-D back trajectories. Note that the scales on the *y* axis vary.

[12] The results, presented in Figure 2, show that for Rigi in Switzerland and Peyrusse Vieille in southern France the residence time in the European PBL was particularly high in June and August. Also at Kosetice in the Czech Republic the residence time in the European PBL was high in June. At the other sites, further north (Mace Head and Waldhof) and east (Starina), the PBL residence times varied during summer, but without a clear perturbation. This is in line with the geographical distribution of the temperature anomalies which show the highest values in south and central France, Switzerland, southern Germany and northern Italy [*Fink et al.*, 2004]. A lengthened residence time over surface emission areas is only one of several parameters necessary for more effective ozone formation. Additional parameters favoring ozone production include more solar radiation (little cloud cover), higher temperatures and slower mixing processes. As mentioned and further discussed below, such conditions persisted during the 2003 heat waves. Thus, the situation was particularly favorable for significant ozone formation for several weeks in south and central parts of Europe, whereas the regions to the north and east were more at the outskirts of the anticyclone and to a higher extent a receiver of photochemically processed air masses.

3. Surface Ozone

[13] Surface ozone measurements have been a part of the EMEP extended measurement activities since its third phase, and the monitoring started in the late 1980s [*Dollard et al.*, 1995; *Hjellbrekke and Solberg*, 2004]. A total of 131 stations in 27 European countries reported data for 2003. The ozone monitoring sites are situated mainly in central, western and northern Europe and the network density is

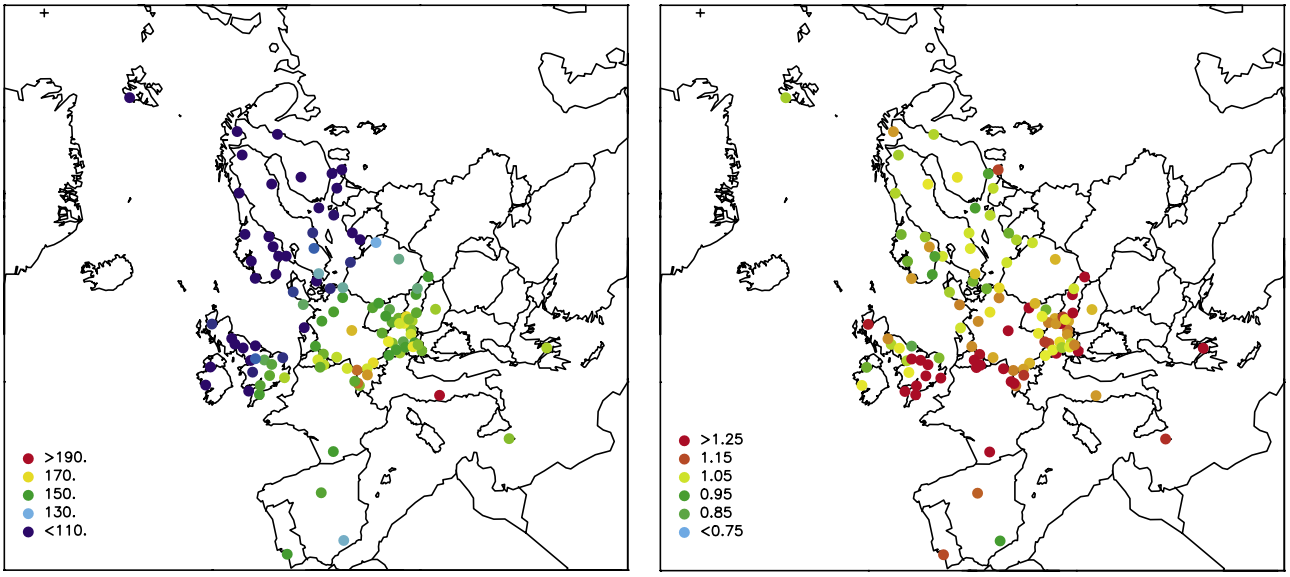

Figure 3. (left) The 95-percentile of daily maximum ozone concentrations observed in 2003 in μg m^{-3} and (right) this 95-percentile relative to the 95-percentile of the daily maximum ozone values observed during the period 1991–2002.

poorer in the eastern and Mediterranean parts of Europe. The stations are located in rural or remote areas, away from local emission sources, and thus representative of the regional concentration field.

[14] In many of the countries there are national or regional networks with a large number of ozone monitoring sites. The dedicated EMEP sites have been selected from these by the countries and by EMEP's Chemical Coordinating Centre (EMEP-CCC) at the Norwegian Institute for Air Research (NILU) on the basis of certain criteria of site location, as specified in detail by the EMEP manual [*EMEP*, 1995]. The most important criteria is that the sites should be regionally representative, thus located away from nearby emission sources, and, second, not located in certain topographic areas like valleys subject to inversion situations or on mountain tops or passes. The monitoring data are subject to a strict quality control procedure as defined in the EMEP manual, both in the individual countries and at the EMEP-CCC, before being accepted as valid data. Information about the ozone data quality, calibration and maintenance procedures was collected from the participants during 2000 [*Aas et al.*, 2001]. The UV-absorption method was the only measurement method in use in 2003.

[15] An upper extreme value of ozone is a better indicator for photochemical episodes than, e.g., a mean value as the latter is to a larger extent controlled by the background values. The 95-percentile ozone concentrations in 2003 (based on daily maximum hourly values) are given in Figure 3. Also given is the ratio of the 95-percentiles in 2003 relative to the 95-percentiles observed during the period 1991–2002. Note that the number of years with ozone monitoring varies between the sites so that the ratios shown in Figure 3 do not refer to the same group of previous years for all sites. The ratios were only calculated for sites with ozone monitoring back to at least 1998. Figure 3 shows that the 95-percentile ozone concentration in 2003 exceeded 160–170 μg m^{-3} over a large region in central Europe extending from Austria in southeast across most of Germany to Belgium, the Netherlands and the southeast part of the UK. The 95-percentiles in 2003 exceeded the previous 95-percentiles in France and at several sites in Switzerland, Germany and Austria. Also in the most northern part of Scandinavia, record-breaking ozone values were observed around 20 April 2003 peaking at 85 ppb (170 μg m^{-3}) at Esrange in northern Sweden as discussed in detail by *Lindskog et al.* [2007].

Table 1. Location, Magnitude, and Date of the 10 Highest Hourly Ozone Concentrations Observed by the EMEP Network in 2003

Station Name	Country	Longitude	Latitude	Maximum Value, μg m^{-3}	Date Observed
Eupen	Belgium	6°00′E	50°38′N	296	8 Aug 2003
Montelibretti	Italy	12°38′E	42°06′N	287	13 Jun 2003
Donon	France	7°08′E	48°30′N	254	11 Aug 2003
Harwell	UK	1°19′W	51°34′N	246	15 Jul 2003
Vreedepeel	Netherlands	5°51′E	51°32′N	244	7 Aug 2003
Schmücke	Germany	10°46′E	50°39′N	243	12 Aug 2003
Vezin	Belgium	4°59′E	50°30′N	239	8 Aug 2003
Revin	France	4°38'E	49°54′N	239	8 Aug 2003
Bassum	Germany	8°43′E	52°51′N	238	12 Aug 2003
Lullington Heath	UK	0°11′E	50°48′N	236	11 Aug 2003

[16] In contrast, the 2003 peak values in UK were lower than the maximum values for previous years except for Wicken Fen, and except for Harwell where the 2003 peak value reached the same peak value, 246 μg m^{-3}, as observed during 1991–2002. The reason for these regional differences is both that the main area of the European ozone plume in 2003 was located south of UK and also that most of the UK sites have continuous monitoring data back to 1991 (and before) compared to, e.g., the French sites which have a shorter monitoring history. It has been estimated that peak ozone concentrations at the EMEP stations in the UK declined about 30% in the period 1986–1999 [*National Expert Group on Transboundary Air Pollution*, 2001]. The main reason for this decline is believed to be the reduction in emissions of ozone precursors in Europe.

[17] The ten highest ozone concentrations observed in the EMEP network in 2003 are given in Table 1. The highest value of 296 μg m^{-3} was observed at Eupen in Belgium. Most of the 10 highest peak values occurred during the first half of August with a few exceptions. At Montelibretti (near Rome) in the south the peak value occurred in June and in UK the peak value at Harwell was seen in mid-July.

[18] Figure 4 shows the monthly means of daily maximum (MDM) ozone values in 2003 for each of the months March–August relative to the highest MDMs during the years 1991–2002. The MDMs in June and particularly August were record high compared to the data back to 1991 over a large region in central Europe. At Payerne and Tänikon in Switzerland the MDM in August 2003 was approximately 15% higher than in any other August since 1991. However, the MDMs were also record high in other periods of 2003, like in March and April in parts of Scandinavia and the UK. This shows that Europe in 2003 experienced record-high ozone concentrations in several individual periods during the whole spring/summer period. In May and July 2003 the MDMs in central Europe and UK were lower than the maximum of the MDMs for previous years at most sites. In middle and northern Scandinavia, however, peak MDM values were observed in July.

[19] Figure 5 shows the seasonal cycle in ozone measured at 8 sites in the EMEP network in 2003 compared to the average seasonal cycle based on the 12-year period 1991–2002. The seasonal cycles were calculated as 30-d running median of the daily maximum of the hourly ozone concentrations. The results shown in Figure 5 indicate a most pronounced perturbation in the ozone level during June–August 2003 at the central European sites (Waldhof, St. Koloman, Schmücke and Payerne) with a deviation from the 12-year reference of the order of 40–50 μg m^{-3} at most. During these periods the running median values for 2003 occasionally exceeded the running 90-percentiles of the reference climatology. In the southern UK, represented by data from Yarner Wood, a marked period of enhanced ozone concentrations are seen in summer, although lower concentrations than those observed at the central European sites. Further north in the UK, e.g., at sites in Scotland, the 2003 data showed only a minor deviation from the reference. At Mace Head, on the west coast of Ireland, slightly elevated mean ozone levels are evident from the middle of May to the middle of August. In July and August 2003 Scandinavia experienced positive monthly temperature anomalies of around 2°C compared to the 1961–1990 climatological average but was located outside of the main area of the heat waves. The ozone measurements from Birkenes (Figure 5) only have a minor peak in August while at Rørvik in southwest Sweden slightly elevated levels are seen from June to the beginning of August.

[20] Several studies have reported a long-term gradual rise in the background ozone concentration [*Simmonds et al.*, 2004; *Laurila et al.*, 2004] and this is of potential relevance when comparing the 2003 data with the reference based on the preceding 12 years of data as there is likely to be a slight trend inherent in this reference. However, our calculations show that this long-term trend is present in the 12-year period, but is small and negligible compared to the large difference between the 2003 data and the 12-year reference.

[21] As mentioned above, it is not surprising that the European heat waves were accompanied by enhanced ozone concentrations. Increased UV radiation, reduction in clouds and precipitation, extended residence time in the polluted boundary layer, very high temperatures, increased biogenic emissions and drought all lead to higher ozone levels. Model calculations by *Vautard et al.* [2005] showed a better model performance when the model was run with reduced dry deposition due to low soil moisture. Furthermore, there is a direct link between temperature and efficiency of ozone formation [*Sillman and Samson*, 1995] particularly important during high temperature situations that may have contributed significantly in the summer 2003. This is caused by a strong thermal destabilization of PAN and other organic nitrates as the temperature increases, increasing the NO_x concentration. NO_x is normally the limiting factor for ozone formation in rural areas. At lower temperatures PAN ties up a significant fraction of the nitrogen oxides [*Sillman and Samson*, 1995] thereby reducing the ozone formation.

[22] Not all meteorological perturbations experienced during heat wave situations constitute positive feedback systems with respect to ozone formation, though. The concentration of the OH radical is closely coupled to the absolute humidity of the atmosphere through the reactions:

$$O_3 + h\nu \rightarrow O_2 + O(^1D) \quad (1)$$

$$H_2O + O(^1D) \rightarrow 2OH \quad (2)$$

which is the main production channel of the OH radical in the troposphere. The OH radical is the main driver of the tropospheric photochemistry and although an ozone molecule is consumed in the reactions given above, the OH radicals will give rise to a net ozone production as long as the NOx level is above a minimum level (typically NO > 10 pptv). In dry air, however, associated with high-pressure systems, the OH production can be reduced compared to a situation with higher, more normal, concentrations of water vapor. This constitutes a negative feedback mechanism between heat waves and ozone formation.

[23] When considering the ozone peak values and maximum in MDM in June and August 2003 as presented above, it should be kept in mind that the emissions of European ozone precursors have been substantially reduced

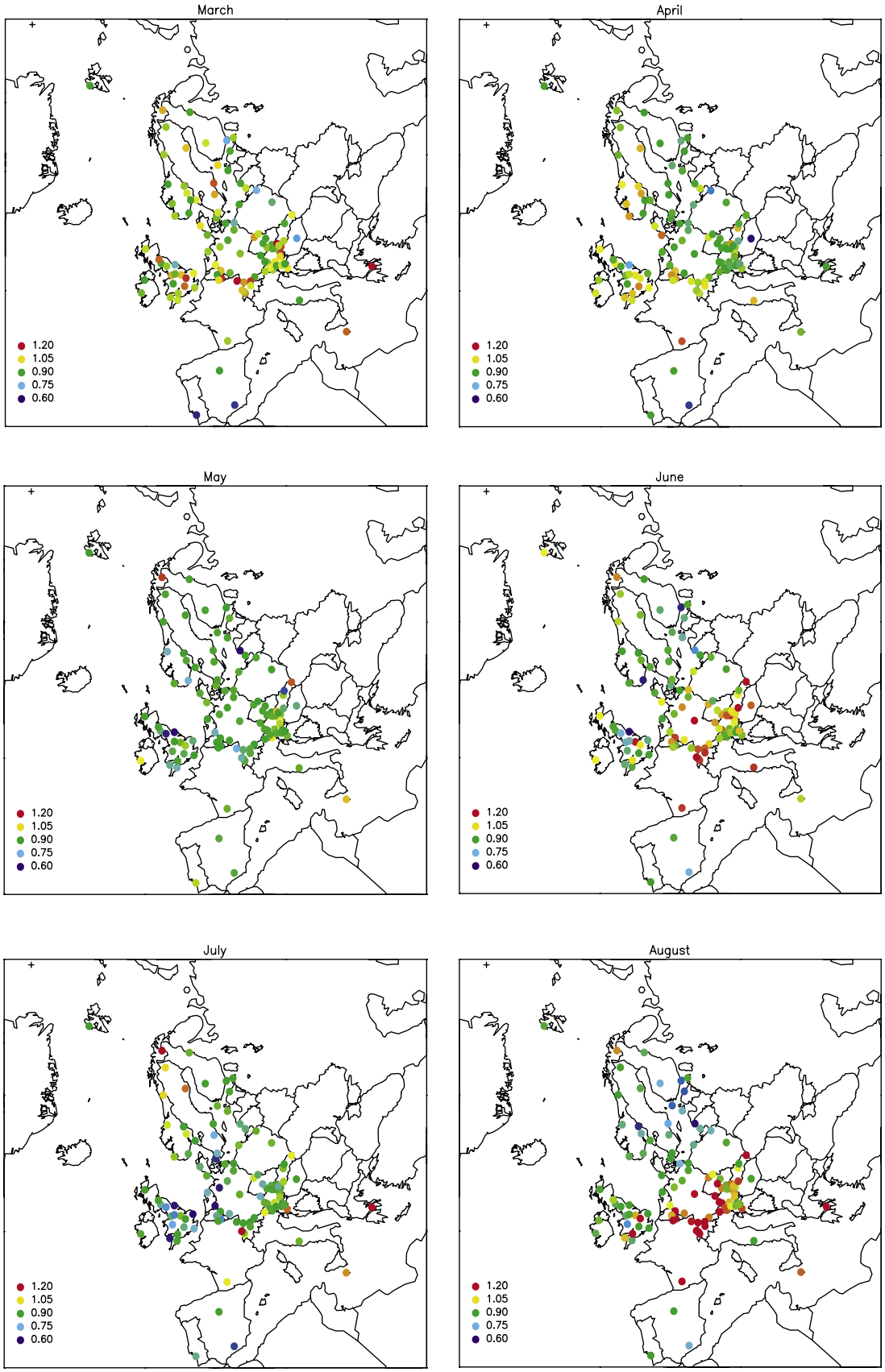

Figure 4. Monthly means of daily maximum ozone concentration observed in 2003 relative to the maximum of the monthly means of daily maximum ozone during the years 1991–2002.

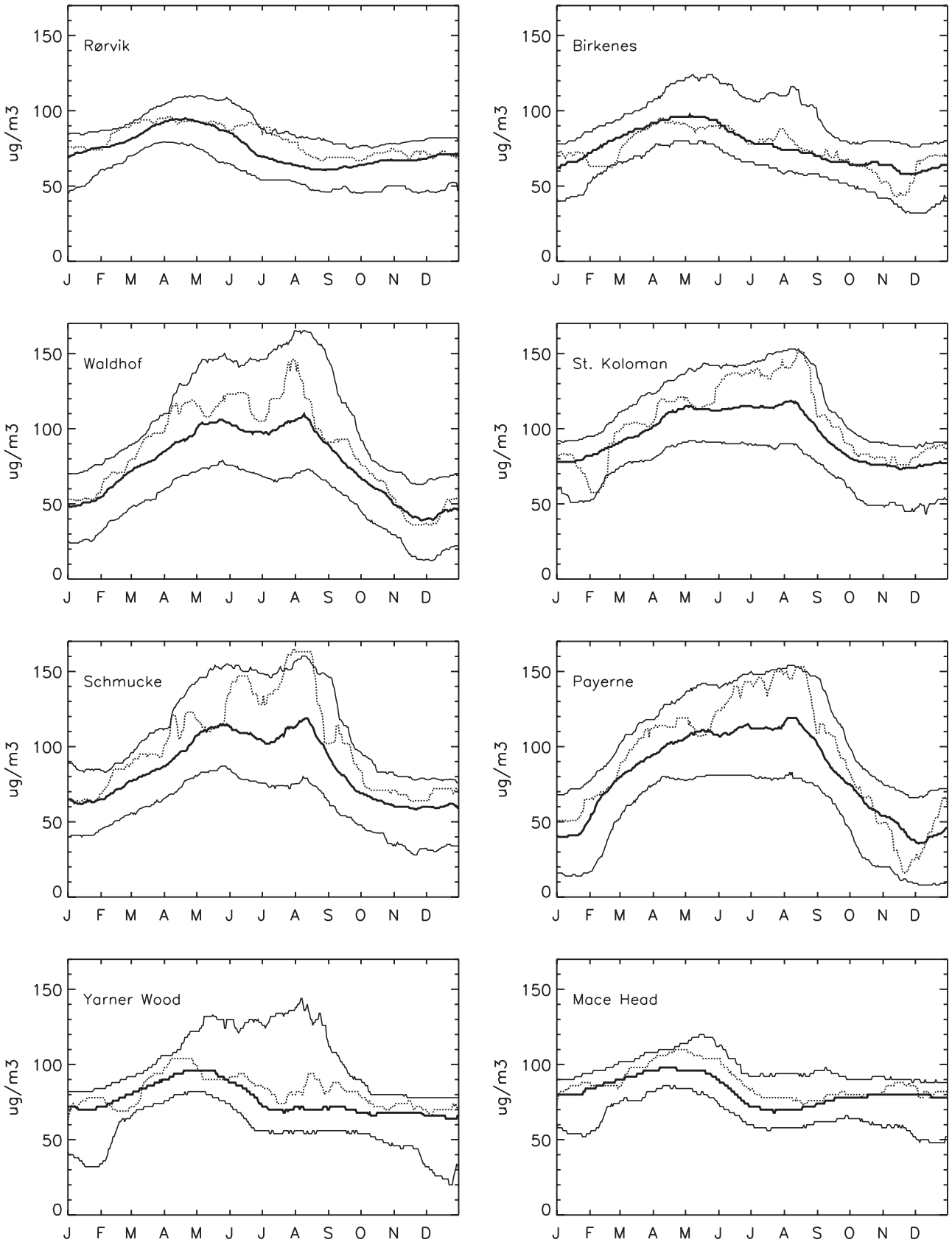

Figure 5. The 30 d running median ozone concentrations for 2003 (dotted) and for the 12-year period 1991–2002 (bold). The 30 d running 10th and 90th percentile ozone concentration for the 12-year period are also given (thin lines). All data are based on daily maximum concentrations in μg m^{-3}.

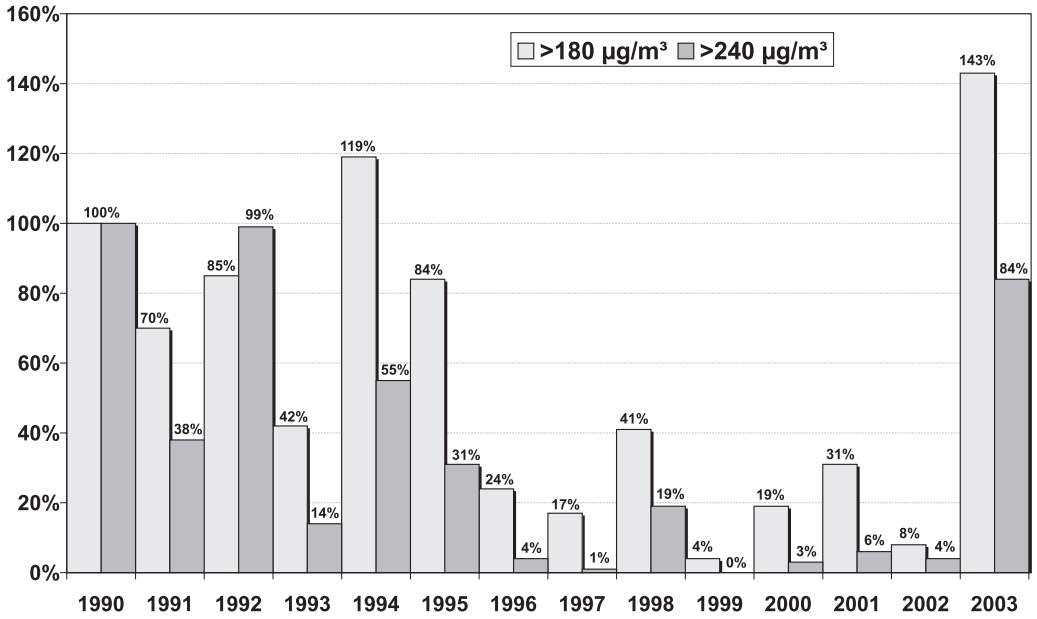

Figure 6. The number of hourly exceedances of the threshold level of 180 μg m^{-3} for all German ozone monitoring sites (EMEP and national) relative to the year 1990 and relative to the number of sites [*Umweltbundesamt*, 2003].

during the last 10–15 years. The annual emissions of NO_x and VOC within the whole EMEP region have been reduced by 23% and 32%, respectively during the period 1991–2002 [*Vestreng et al.*, 2004], and the emission reductions in west and central Europe are even larger. According to *Vestreng et al.* [2004] the emission reduction of NO_x in Germany and France during this period has been 43% and 31%, respectively, and the reductions of VOC emissions have been even larger. In spite of the substantial reduction in German NO_x and VOC emissions the number of exceedances of the 180 μg m^{-3} was record high in 2003 as seen in Figure 6 [*Umweltbundesamt*, 2003]. Such increase in the peak ozone concentrations should be seen in the context of health damage as given, e.g., by EU's threshold levels of 180 and 240 μg m^{-3}. The threshold level of 180 μg m^{-3} is the so-called information threshold, defined as a level beyond which there is a risk to human health from brief exposure for particularly sensitive sections of the population. The 240 μg m^{-3} is the so-called alert threshold, defined as a level beyond which there is a risk to human health from brief exposure for the general population.

[24] Figure 7 shows the wind and temperature on the 850 hPa surface together with the daily maximum ozone concentrations observed at the EMEP network stations during the August heat wave. In Portugal and Spain the winds were southerly, gradually weakening through the period, whereas in northwest Europe westerly and southwesterly winds were dominating and in northeastern Europe the winds were northwesterly, associated with the anticyclone located over central Europe. At the beginning of the period a tongue of warm air gradually protruded from Spain and Portugal and to the northeast, whereas in the middle of the period, stable and high temperatures were found over a large region from Portugal to France, Switzerland, southern Germany and northern Italy. Extreme temperatures exceeding 24°C at the 850 hPa surface (1.5–2 km above sea level) were found in southwest Europe.

[25] At the beginning of the August period the highest surface ozone concentrations were seen in central Germany, eastern France and the southern part of the BeNeLux region. This was northeast of the area of inflow of warm air masses from southwest, whereas in Spain the ozone concentrations were only slightly elevated. Later in the period the area of maximum ozone concentrations coincided with the area of particularly high temperatures in the 850 hPa surface, and also coinciding with low wind. At the end of the period, from 12 August and onward, the warm and ozone-rich air masses moved to the southeast as a cold front gradually approached from the northwest Atlantic Ocean.

[26] The potential temperature and ozone concentration between the surface and 5 km as measured by soundings on 4, 6, 8, 11 and 13 August in Uccle, Belgium, are shown in Figure 8. In the first part of the period (4 and 6 August) the elevated ozone concentrations were confined to a fairly shallow planetary boundary layer (PBL) of 0.5–1 km depth. From 6 to 8 August there was a substantial shift in the vertical ozone distribution with very high mixing ratios exceeding 100 ppbv from the top of the PBL to 2.5 km altitude, whereas within the PBL the mixing ratios were of the order of 60–80 ppbv. On 11 August the sounding indicate elevated mixing ratios of the order of 80–90 ppbv throughout the lowest 5 km, whereas on 13 August this was reduced to more normal values of 40–50 ppbv, except for a layer of moderately enhanced ozone between 1 and 1.5 km.

[27] The depth of the PBL, as indicated by the potential temperature curves, was 0.5–1.5 km and capped by a very stable stratification during these days. On 4 and 6 August the vertical ozone profiles matched the profiles for the potential temperatures, indicating local photochemical ozone formation in the PBL. The strong ozone enhancement between 1 and 2.5 km on 8 August was located above a

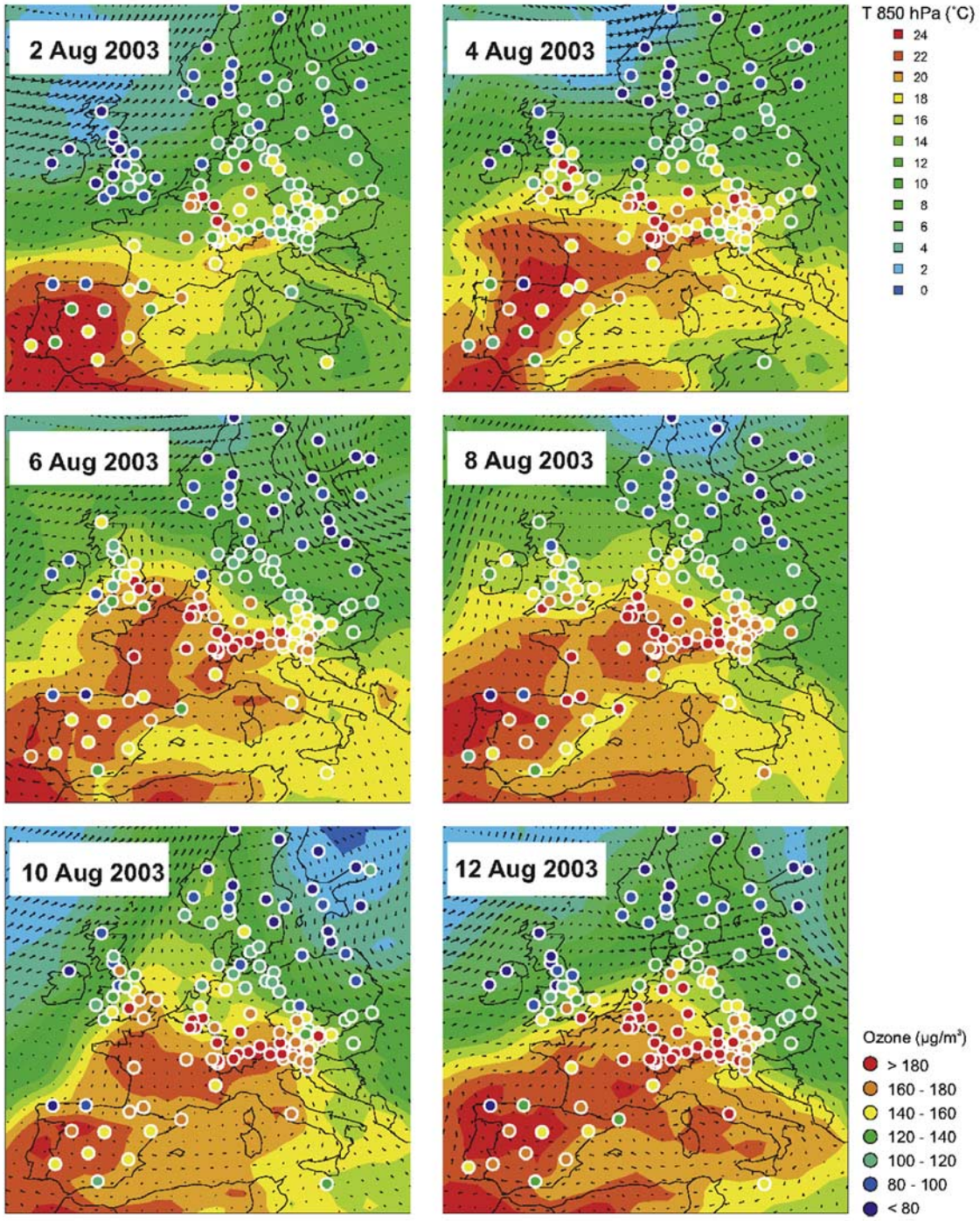

Figure 7. The daily maximum ozone concentrations (solid circles) on top of the temperature on the 850 hPa surface (°C) and the wind at the same level during the August 2003 episode.

very stable PBL, making local vertical transport of ozone or ozone precursors from the surface unlikely. Rather this suggests advection above the PBL of an air mass enriched in ozone. The subsequent increase in ozone in the PBL and above 2.5 km on 11 August can be explained by a gradual downward and upward mixing of this ozone-rich layer. The rise in the height of the PBL from 8 to 11 August supports this explanation.

[28] Sensitivity runs with the chemical transport model at the University of Oslo ("Oslo CTM2") for August 2003 were carried out to estimate the importance of a number of processes for the high ozone levels observed. A description of the model and several applications are given by *Brunner et al.* [2003, 2005], *Isaksen et al.* [2003, 2005], and *Gauss et al.* [2006]. The Oslo CTM2 is a global chemical transport model, with a detailed chemical description of the tropo-

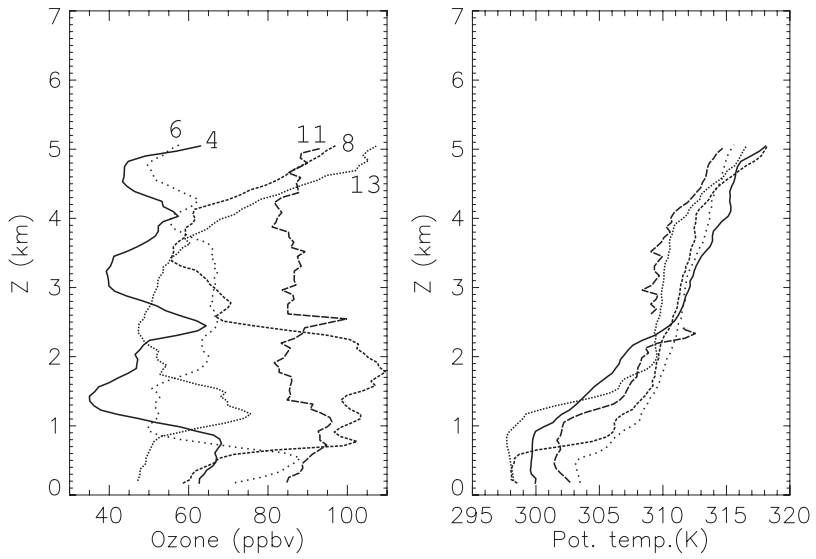

Figure 8. Measurements of (left) ozone mixing ratio and (right) potential temperature by five soundings released at Uccle, Belgium, in August 2003. The numbers on top give the days the sondes were launched.

sphere and the stratosphere. It is driven by ECMWF integrated forecast system model (operational forecast) meteorology. The horizontal resolution is T42 (2.8 × 2.8 degrees) and the vertical resolution is 40 layers extending from the surface up to 2 hPa. The simulations were done for three months using emissions corresponding to year 2000. This model run was initiated by running the model for three months in 2003 (1 April to 1 July), and this initialization run was in turn initialized by a previous modeling result for the monthly mean of April 2005. From experience three months is sufficient time for this model to reach correct levels when started by a monthly mean concentration from another year. Hourly ozone mixing ratios in the lowest model grid box which extends from the surface to 16 m were compared to observations at several European locations.

[29] In addition to a standard model run, five sensitivity runs were performed, where model parameters were modified in a subdomain confined by 10°E to 15.5°W and 36°N

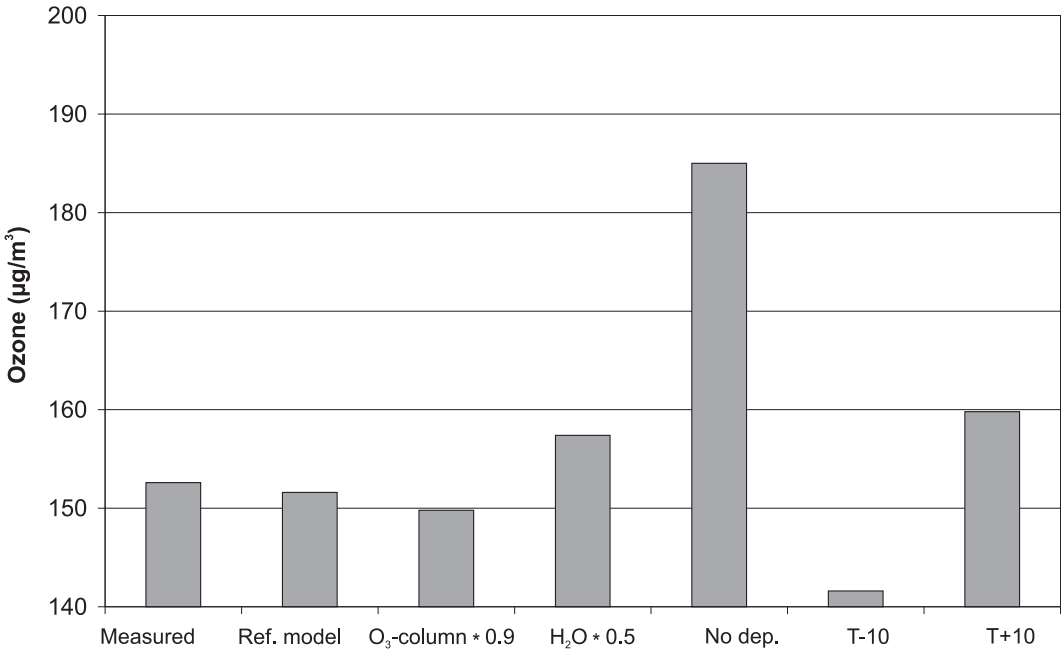

Figure 9. Average maximum hourly ozone concentration at eight stations on 8 August 2003 as measured and calculated in the reference model run with the Oslo CTM2 and in sensitivity runs with the ozone column reduced by 10%, water vapor reduced by 50%, zero surface deposition, and the temperature decreased and increased by 10°C inside the subdomain confined by 10°E to 15.5°W and 36°N to 56°N and below 3.5 km height.

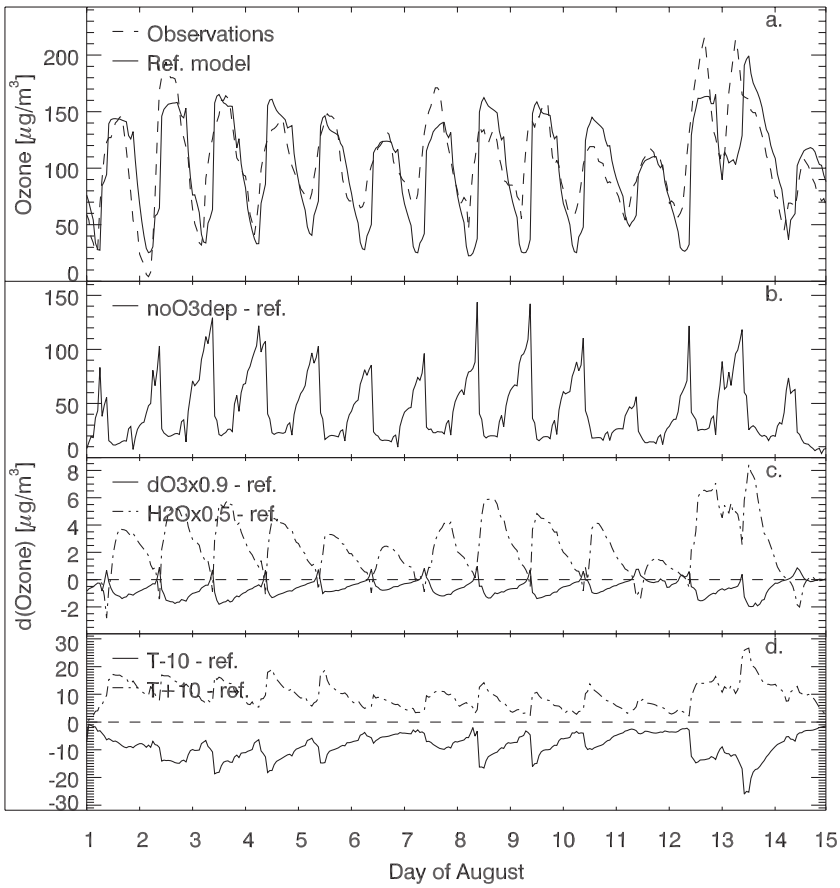

Figure 10. (a) Surface O_3 concentrations as observed and modeled with the Oslo CTM2 for EMEP site Waldhof in north Germany during 1–15 August 2003 as well as (b) the calculated perturbation in O_3 without dry deposition, (c) the total ozone column reduced by 10%, water vapor concentration halved, and (d) with temperature changed ±10°C in the chemistry part of the model calculation.

to 56°N as follows: (1) zero surface dry deposition, (2) stratospheric ozone column reduced by 10% [*Orsolini and Nikulin*, 2006], (3) 50% reduction in absolute humidity below 3.5 km, (4) 10°C increase in temperature for chemical reactions below 3.5 km, and (5) 10°C decrease in temperature for chemical reactions below 3.5 km. The changes chosen in these parameters correspond to what can happen as the land surface is drying out in an intense heat wave like the one in the summer 2003.

[30] Figure 9 shows the average of the daily maximum in the hourly ozone concentration calculated for 8 stations in central Europe on 8 August, at the peak of the episode. The stations Donon (FR), Revin (FR), Morvan (FR), Peyrusse Vieille (FR), Campisabalos (ES), Kosetice (CZ), Waldhof (DE) and Zingst (DE) were used. The time series of observed and modeled ozone together with the changes calculated by the sensitivity runs are presented for Waldhof in north Germany in Figure 10. The observed diurnal variation is usually reproduced quite well. The maximum concentrations are in reasonable agreement at some sites, less so at others. It should be mentioned, however, that the purpose of these calculations were not to carry out an extensive model validation study. The purpose of this exercise was to quantify the magnitude of influence of these perturbations and how they ranked in importance as judged by the change in the ozone concentration with the aim to estimate the strength and sign of major feedback mechanisms linked to the meteorological situation in August 2003 on surface ozone. These feedback mechanisms are all at work in a weather situation like the August 2003 heat wave. In drought, vegetational uptake of ozone may be reduced or even stopped because of the plants closing their stomata. Also, high-pressure situations are typically accompanied by an elevated tropopause height and a reduced ozone column leading to increased UV radiation to the troposphere, and increasing the UV-dependent photolysis rate of in particular the O_3 photolysis to $O(^1D)$. A reduction in the water vapor content and enhanced temperatures are typically experienced in this kind of meteorological situations.

[31] The Oslo CTM2 model results are shown in Figure 9 for one day only. These results turned out to be well representative for the whole period 1–14 August, however, although there were some differences among the individual sites. Turning off the dry deposition inside the subdomain confined by 10°E to 15.5°W and 36°N to 56°N had the strongest effect on ozone and resulted in an increase in the mean daily maximum hourly mixing ratio of 34 μg m^{-3} compared to the reference calculation of 152 μg m^{-3}, an increase of more than 20%. Turning off the dry deposition altogether is not physically realistic, not least because there

Figure 11. Picture from the Terra satellite on 4 August 2003, 1130 UTC, showing extensive fires (marked with red symbols) and smoke plumes moving north on the western part of the Iberian Peninsula. Image courtesy of MODIS Rapid Response Project at NASA/GSFC.

is a significant nonstomatal deposition flux, but it shows the upper limit of the impact of this process. Dry removal is the dominant loss mechanism for ground-level ozone. Next, changing the temperatures inside the subdomain and in the chemistry part of the model calculation only by ±10°C ranked as number two in influence on ozone with a calculated change of the order of ±8 μg m^{-3} ozone, corresponding to 5% of the ozone mixing ratio in the reference calculation. Thus, we conclude that the high temperatures in August 2003 contributed of the order of 5% to the high ozone concentrations calculated, through a more efficient photochemical formation. A 50% reduction in the atmospheric content of water vapor caused the ozone concentration to increase by 2–3%. Water vapor can influence the photochemistry in several ways, e.g., by reducing the OH formation (through the reaction $H_2O + O(^1D) \rightarrow 2OH$) reducing the rate of the VOC oxidation and thereby less ozone formation, and by increasing the lifetime of NO_x with respect to OH (through the reaction $NO_2 + OH \rightarrow HNO_3$) thereby allowing more ozone formation per NO_x molecule. The small effect on ozone of a 50% reduction in H_2O can be explained by these two counteracting effects. Finally, the effect of a reduced total ozone column, thereby giving rise to an increase in the photolysis $O_3 \rightarrow O(^1D)$ followed by increased OH formation, was negligible when the ozone column was reduced by 10%.

4. Forest Fires

[32] There were forest fires in many parts of Europe, particularly in the south, and most pronounced in Portugal and Spain in the summer of 2003. The Iberian Peninsula experienced periods of extensive forest fires during July to September. In the first half of August, coinciding with the peak heat wave in central and northern Europe, massive fires were observed in Portugal, as seen from the Terra satellite 4 August (Figure 11). Long-range transport of fire emissions have been found to give rise to significantly elevated ozone concentration. For example events of transport of biomass burning from North America to the Azores leading to an ozone increase up to 30 ppbv have been identified [*Honrath et al.*, 2004].

[33] To look for a possible link between the Portuguese forest fires and the ozone episode further north, backward simulations were carried out with the Lagrangian particle dispersion model FLEXPART [*Stohl et al.*, 1998] carried out in the same way as described by *Seibert and Frank* [2004]. The FLEXPART model does not calculate any chemistry but is extremely useful for calculating the origin of air masses and atmospheric transport pathways. It is superior to single-trajectory computations. FLEXPART parameterizes turbulence [*Stohl and Thomson*, 1999] and convection [*Emanuel and Zivkovic-Rothman*, 1999; *Forster et al.*, 2007], and was validated in a number of studies on air pollution transport [e.g., *Wotawa and Trainer*, 2000; *Forster et al.*, 2001; *Spichtinger et al.*, 2001; *Stohl et al.*, 2003]. The simulations for this study are based on operational data (horizontal resolution of 1° × 1°, 61 vertical levels, temporal resolution of 3 h) from the *European Centre for Medium-Range Weather Forecasts* [1995]. At the (receptor) site Eupen, Belgium, the site where the highest ozone value within the EMEP network was measured (Table 1), 20 000 particles with unit mixing ratio were released every 6-h interval between 1 and 12 August and followed 20 d backward in time. A response function to emission input which is related to the particles' residence time and can be used to determine the source regions and pathways of air masses to the receptor, was then calculated on a uniform grid.

[34] Figure 12 shows the sum of the total columns of the sensitivity function over the last 3 to 20 d before arrival on 7 August between 0000 and 0600 UTC at Eupen. The air masses originate mainly from the Bay of Biscay and the western parts of the Iberian Peninsula, close to where the forest fires burned during the first half of August. The values of the response function are also high over northern Germany and parts of the Netherlands indicating that the air at Eupen originate partly near by and partly further away. Similar patterns in the source regions and the pathways of air masses to Eupen were found for 8 to 12 August (not shown), whereas before 6 August transport from the Atlantic and North America dominated. A closer look at the FLEXPART results indicates a transport time from the Iberian Peninsula of around 4 d. These results indicate that the forest fires in Portugal/Spain during August could indeed have contributed to the peak values in surface ozone observed in north Europe.

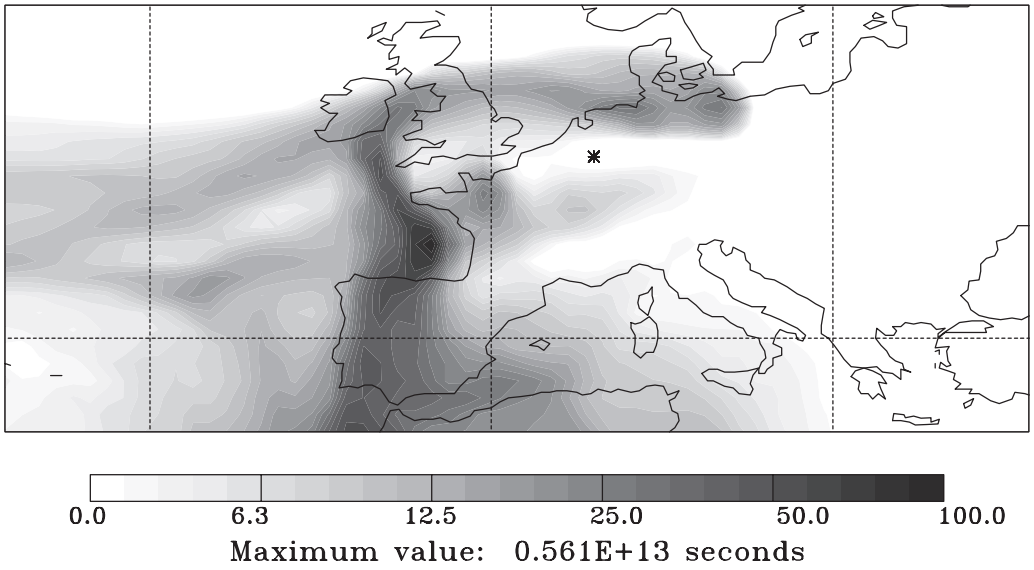

Figure 12. Total columns of the response function to emission input (indicative of the residence time) over the last 3 to 20 d of all particles arriving at Eupen on 7 August between 0000 UTC and 0600 UTC on a $1^\circ \times 1^\circ$ grid as calculated with FLEXPART. The residence times are given in relative units of the maximum residence time below the panel.

[35] The results from the FLEXPART model agree with the development of the synoptic situation as shown in Figure 7 and as discussed above. In the beginning of the period elevated ozone concentrations in central Europe were likely caused by local formation in the PBL, whereas later in the episode, advection of ozone-rich, warm air masses from southwest above the PBL was more likely. Thus, as the area of forest fires and the influence region for the air masses arriving at Eupen are similar, it is likely that these forest fires contributed to the enhanced ozone concentrations observed. To quantify the influence, however, requires photochemical modeling on a fine scale together with high-resolution spatially and temporally resolved data on the emissions from the fires, and this is beyond the scope of the paper.

5. Isoprene

[36] Isoprene has been measured at the Donon monitoring site (48°30'N, 7°08'E) in southeast France since 1997 as part of the EMEP VOC monitoring programme [*Borbon et al.*, 2004; *Solberg*, 2005]. The samples are collected in electro polished stainless steel canisters twice a week and subsequently analyzed by GC/FID for about twenty C_2–C_7 individual light hydrocarbons by the laboratory of Ecole des Mines des Douai (EMD) in France. Figure 13 shows the

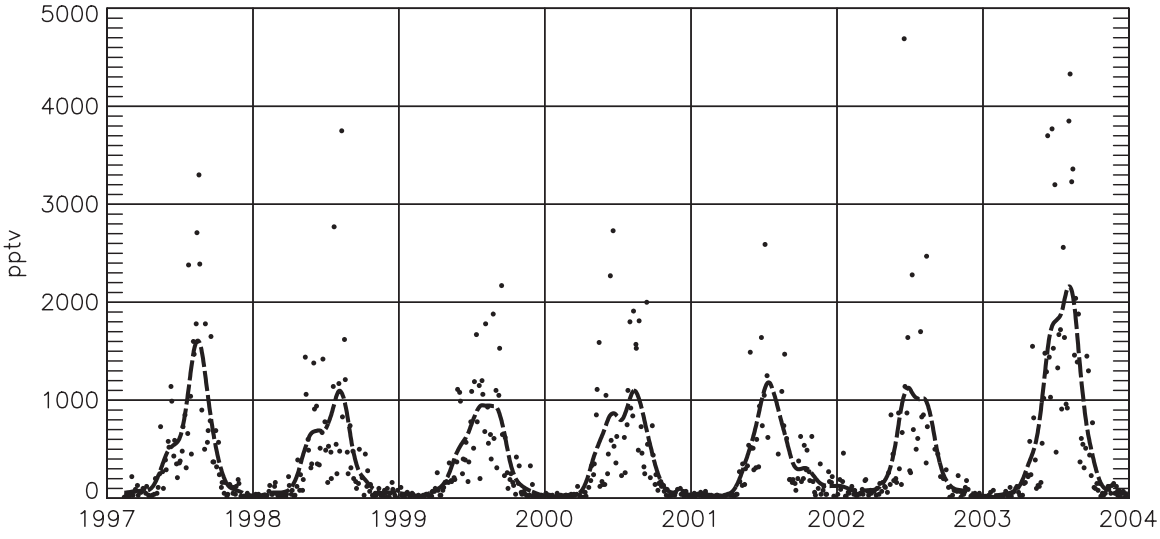

Figure 13. EMEP canister samples of isoprene at Donon during 1997–2003. Individual samples (twice a week) are shown as marks and the corresponding smoothed running average as a dashed curve.

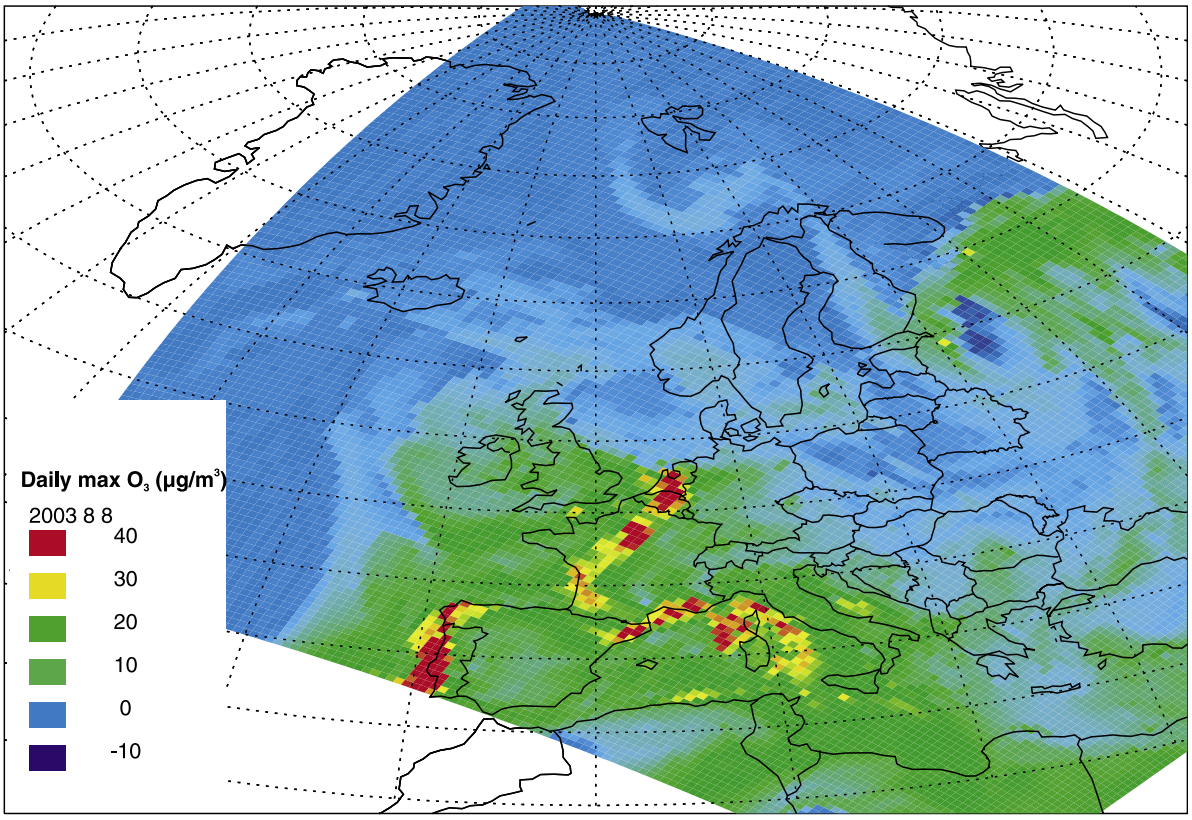

Figure 14. The change in max surface ozone (μg m^{-3}) on 8 August 2003 due to biogenic isoprene emissions as calculated with the EMEP unified model.

measured isoprene concentrations from 1997 to 2003 at Donon. Whereas the running average mixing ratio peaked at approx. 1 ppb in the summers of 1998–2002, the concentration level in summer 2003 was about twice this value. Also in summer 1997 the mean isoprene level was elevated but not as much as in 2003.

[37] Biogenic isoprene emissions are strongly controlled by solar radiation and surface temperatures [*Guenther et al.*, 1993; *Simpson et al.*, 1999]. One explanation for the isoprene concentrations observed at Donon in 2003 is increased biogenic emissions coinciding with the elevated temperature and solar radiation during summer 2003. In a previous study of this station, *Borbon et al.* [2004] concluded that in summer, in situ biogenic emissions constitute at least 80% of isoprene, whereas, in winter, more than 90% of residual rural isoprene comes from urban air mass mixing. Additionally, the surface isoprene concentrations could be enhanced compared to normal summer conditions because of reduced vertical mixing associated with stable, anticyclonic conditions. Thus, without micrometeorological measurement data and local-scale modeling, it is difficult to distinguish between these two effects. In periods with severe drought (low soil moisture content) and high temperatures the plants regulate their water loss by decreasing the stomata opening. However, as discussed by *Niinemets and Reichstein* [2003] the reduction in stomata during periods of drought giving less water transpiration and less ozone uptake, is normally not affecting the emission of isoprene and α-pinene. We also note that isoprene was measured in 2003 at a few other sites in Germany and the Czech Republic and the concentrations there were not elevated [*Solberg*, 2005]. This could be explained by the fact that these other sites are less exposed to isoprene emissions, whereas Donon, located near the forest is more indicative of the direct biogenic emissions.

[38] Furthermore, the effect of elevated isoprene concentrations on the ozone formation depends on the NO_x-concentration level and whether the ozone formation is NO_x- or VOC-limited. Previous calculations with the Lagrangian EMEP oxidant model [*Simpson*, 1995] indicated that mean ozone levels in Europe were fairly insensitive to isoprene as the major part of the isoprene emission was in NO_x-limited regions, whereas in VOC-limited areas anthropogenic VOCs dominated over biogenic VOCs. For peak ozone values, however, the effect of isoprene emissions was calculated to be larger.

[39] To make an estimate of the contribution from biogenic isoprene to the peak ozone levels observed, the EMEP Unified Model [*Simpson et al.*, 2003, 2007] was run for the period 1 July to 31 August 2003 with and without the isoprene emissions, respectively. The EMEP model is a 3D Eulerian atmospheric dispersion model for simulating the long-range transport of air pollution over several years. The model has 20 vertical layers in σ coordinates and is primarily intended for use with a horizontal resolution of $\sim$50 $\times$ 50 km^2 (at 60°N) in the EMEP polar stereographic grid. The chemical scheme uses about 140 reactions between 70 species. The model uses a coupled isoprene

emission algorithm where the emission rates were determined at every time step from the local temperature and global radiation used by the model. The EMEP model was used for this modeling as it has a more sophisticated parameterization of isoprene emission and oxidation and also a finer grid resolution, particularly important for the short-lived isoprene, compared to the Oslo CTM2 model.

[40] Figure 14 shows the contribution of the isoprene emissions to the daily max ozone on 8 August which was at the peak of the episode. This calculation indicates a significant contribution from biogenic emissions to the surface ozone levels, more than 40 μg m^{-3} in some areas in west Europe, corresponding to around 20% of the observed maximum hourly ozone concentration. The calculations thus indicate that in this regional ozone episode a significant fraction of the peak ozone levels could be ascribed to enhanced biogenic emissions. This constitutes an important feedback mechanism relevant for the discussion of what happens to the concentration of ozone in a warmer climate, and indicate that the positive effects of reduced emissions of ozone precursors in Europe (NO_x and VOC) may gradually be outweighed by an increase in the frequency of hot and sunny summer situations [*Solberg et al.*, 2005]. The increase in isoprene contribution to the ozone episode in August 2003 is in line with findings from the TORCH campaign in the UK [*Lee et al.*, 2006]. They found an increase in the overall OH reactivity from isoprene during the episode and also found a strongly nonlinear relationship between measured isoprene and temperature.

6. Summary of Results

[41] The extreme heat waves and drought that Europe experienced in 2003 were accompanied by record-high surface ozone values in central Europe. The highest peak values were for most of the sites observed in August 2003 when the most intense heat wave occurred. The weather development during the August episode, indicate local formation of ozone in the boundary layer during the first days, and advection of warm and ozone-rich air masses from southwest above the boundary layer, which was gradually mixed down to the surface in the subsequent days.

[42] High ozone concentrations prevailed in the summer of 2003 despite of the reduction in the emissions of anthropogenic ozone precursors (NO_x and VOC) in Europe since the 1990s. The unusual meteorological conditions favored a faster formation and a slower loss of ozone than usual. The residence time of air parcels in the European boundary layer was prolonged and there were fewer clouds. Massive forest fires in Portugal and Spain were triggered by the drought and heat, and probably contributed to the ozone peak values in northern Europe in August 2003. Isoprene concentrations were enhanced compared to the seasonal average at a forested site in southeast France indicating either increased biogenic emissions in this area or reduced mixing. Depending on the level of NO_x this could have contributed further to the ozone formation. Model calculations indicated a significant potential contribution from biogenic isoprene to the high ozone concentrations observed, of the order of 20% of the peak ozone values.

[43] A set of sensitivity model runs with the Oslo CTM2 was analyzed to estimate the range of the likely influence on ozone of a number of processes which are affected by the meteorological situation experienced during the heat wave, and which therefore may contribute to surface ozone changes in the future as heat waves of the summer 2003 type may become more prevalent. On the basis of these results we found that the surface dry deposition was a critical parameter, and that reduced deposition due to less uptake through plants may have contributed significantly to the high ozone concentrations in 2003. The upper limit of this effect, calculated by turning the dry deposition off completely inside the subdomain confined by 10°E to 15.5°W and 36°N to 56°N, lead to an increase in peak ozone levels of more than 20%. Second, the feedback between increased temperature and increased photochemical formation was also significant. A 10°C increase in temperature was calculated to give rise to a 5% increase in the peak ozone values during August 2003. The possible influences of modified atmospheric water vapor content or reduced total ozone column were found to be of less importance for the surface ozone concentrations.

[44] Thus, the 2003 summer is a ''field example'' of the close link between meteorological conditions and a secondary pollutant like ozone. Climate model scenarios have indicated that extreme weather events like this may become more frequent in the future. The effect of future climate change may gradually outweigh the benefit of the emission abatement in Europe for summertime secondary photochemical pollutants.

[45] **Acknowledgments.** Meteorological data for FLEXTRA and FLEXPART were provided by ECMWF through the Norwegian Meteorological Institute. The TERRA satellite picture was retrieved from the Internet by courtesy of the MODIS Rapid Response Project at NASA/GSFC. This study was sponsored by EMEP (European Monitoring and Evaluation Programme) and the European Commission through the projects NEPAP (Network for the support of European Policies on Air Pollution), EVK2-CT-2002-80019 and EUCAARI (European Integrated project on Aerosol Cloud Climate and Air Quality interactions), European Commission contract 036833-2.

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