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## A synthesis of the Air Pollution Over the Paris Region (ESQUIF) field campaign

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[1] Tropospheric photooxidant pollution was investigated in detail for the first time over the Paris area during the Air Pollution Over the Paris Region (ESQUIF) project. From 1998 to 2000, 12 intensive observation periods (IOPs) were carried out. They represented various meteorological situations, all leading to strong polluted events over Paris and its surroundings. During these periods, measurements were performed with a new strategy of circular flights around the city, coupled to stations or remote sensing surface measurements. Such data obtained at various altitudes and at different ranges from the city center document the evolution of pollution events on horizontal and vertical scales. In addition, ESQUIF also allowed for the evaluation of models developed in parallel to the project. In this overview, ESQUIF is presented in terms of the set of IOPs. Periods are compared in terms of meteorology and resulting types of pollution episodes. The occurrence of these latter events is discussed in terms of local production and influence of long-range transport. Using both measurements and model simulations, some important results are highlighted, especially concerning accuracy of boundary conditions, processes of mixing within the boundary layer, surface emissions estimation (including biogenic), and photolysis attenuation. Finally, results from data assimilation studies and sensitivity studies using adjoint modeling and a Monte Carlo approach are also presented. *INDEX*

*TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 3210 Mathematical Geophysics: Modeling; 3307 Meteorology and Atmospheric Dynamics: Boundary layer processes; *KEYWORDS:* field experiment, photooxidant pollution, urban- and regional-scale ozone pollution, aircraft measurements

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### 1. General Aim of the ESQUIF Project

[2] Despite progress over the past 20 years in understanding the physics and chemistry that govern air quality, there are a number of unsolved issues in this field. A lack of extensive and continuous observations is partly to blame for

this situation. Although there are routine observations of air quality in some areas, such observations are typically made at ground level and cover a limited set of species. Thus atmospheric chemists have relied on intensive measurement campaigns to address the most pressing questions.

[3] The main goal of the Air Pollution Over the Paris Region (ESQUIF) project was to characterize pollution in a large urban area and to test the ability of air quality models to reproduce the most important features of the urban plume. The campaign involved extensive measurements in the Paris, France, region (Figure 1), a site selected for several reasons which allow the major focus to be on chemical issues. First, Paris is distant from the ocean and from mountains such that the effects of variable orography are minor and meteorological parameters should be straightforward to simulate. Second, it is one of the greatest urbanized areas in Europe, and it is located far from other big cities so that the signatures and origins of pollution are easier to determine.

[4] During ESQUIF, measurements were performed mainly during the summers of 1998 and 1999 in a dozen

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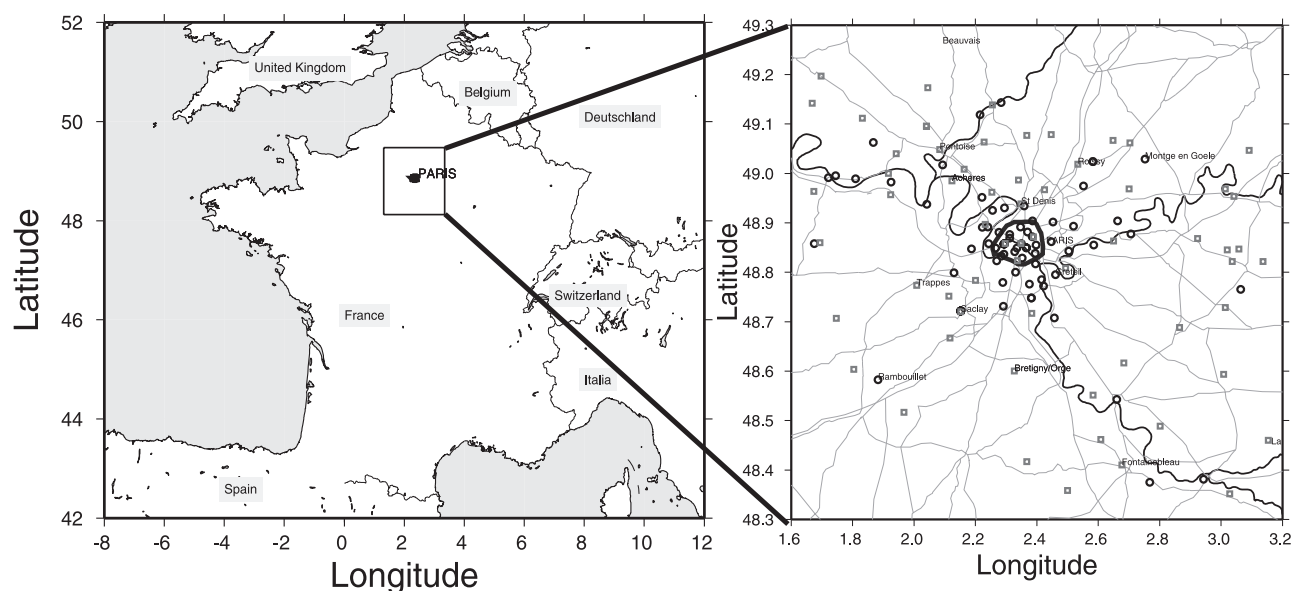
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**Figure 1.** (left) Map of France. The Paris area is shown in the box. (right) Close-up of the Paris area, displaying the locations of the AIRPARIF and Météo France networks as circles and squares, respectively. See color version of this figure in the HTML.

1- to 3-day-long intensive observation periods (IOPs). The instrumental setup is extensively described by *Menut et al.* [2000] and *Vautard et al.* [2001]. The goal of this overview is to highlight the main results obtained during the project. Additional details can be found in subsequent studies of this special section, most of which focus on subsets of results obtained during individual IOPs. We present here an overview of these IOPs. Because results discussed in the following studies deal mostly with photochemistry (i.e., chemical regimes, sensitivity to emissions, etc.) and not with meteorology, here we will present an overview of the main meteorological situations during the campaign.

[5] Prior to the field campaign the ESQUIF scientific committee raised a number of key scientific points that would be addressed by the campaign and during subsequent numerical experiments. The main issues that were addressed in detail are: (1) the sensitivity of ozone formation to a number of factors, including long-range transport of ozone and its precursors from remote, but high emissions, areas in Europe, urban emissions and, specifically, the nature of various chemical regimes, and the importance of biogenic versus anthropic emissions; (2) the evaluation of the available emission inventory, including  $\text{NO}_x$ , CO, and detailed volatile organic compounds (VOCs); (3) the role of aerosols and clouds in the modification of photolysis rates; (4) the role of photochemistry in wintertime nitrogen dioxide episodes; (5) the development of methods and models allowing accurate, spatially distributed representations and forecasts of pollutant fields; and (6) the characterization of aerosol distributions and their evolution around the city of Paris. Some of these points are detailed in the articles of this special section, while others remain under investigation.

[6] Paris is typically not a very polluted city. This is due mostly to the dilution of the urban plume by relatively clean oceanic air and to significant dispersion by prevailing winds. In order to analyze clear signals from urban pollution, we had to focus on the specific, but rare, occasions

when prevailing winds were very weak. Thus a successful IOP required the availability of accurate weather and chemical forecasts several days in advance.

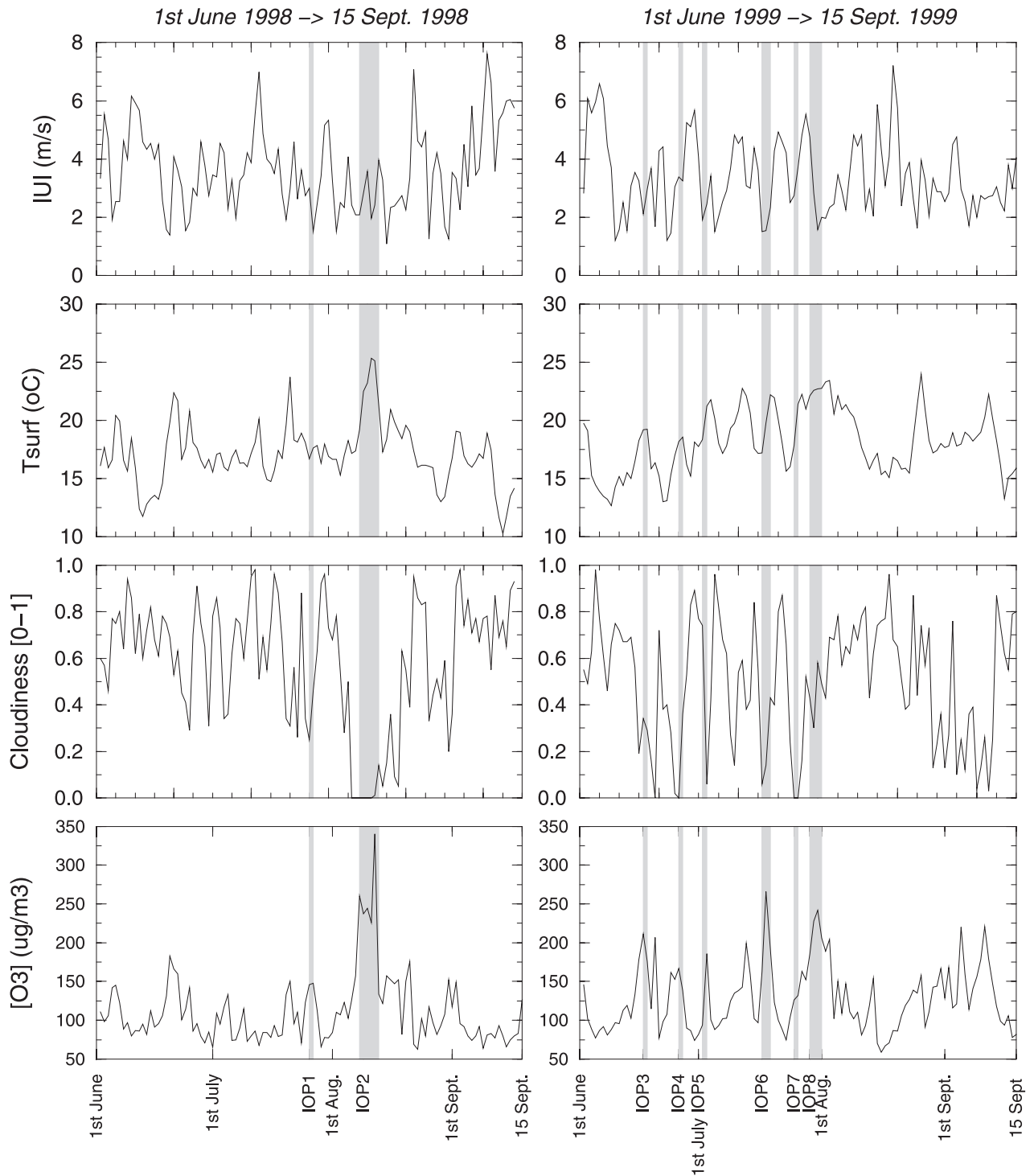
[7] Weather forecasts with specific attention to this campaign were provided by Météo France. Chemical forecasts were performed using several approaches. At the beginning of the campaign (e.g., 1998–1999), operational forecasts of ozone and  $\text{NO}_2$  were only available from statistical models operated by the regional air quality monitoring network AIRPARIF. At that time, several chemistry transport models (CTMs) had also been developed, but they were not fully validated. Thus ESQUIF provided a unique opportunity to test and validate CTMs in forecast modes. In fact, only the CHIMERE model was ready to do such a forecast exercise at that time, and those results are reported by *Vautard et al.* [2001].

[8] The main results of the ESQUIF project are presented in this overview. To highlight IOPs from the summers of 1998 and 1999, the main meteorological characteristics of these summers are presented. To appreciate their differences, characteristics of the various IOPs are compared in section 2. Finally, the research topics studied during the project are described. The major new findings are outlined, with emphasis on those that are described in more detail in the studies that follow.

## 2. Meteorological Conditions During the Summers of 1998 and 1999

### 2.1. IOPs Within the Whole Summers

[9] Figure 2 presents a synthesis of dynamical and chemical conditions that occurred during the summers of 1998 (Figure 2 (left)) and 1999 (Figure 2 (right)). Within each plot, IOPs are shaded (see also Table 1). For wind speed  $|U|(\text{m s}^{-1})$ , surface temperature ( $^{\circ}\text{C}$ ), and cloudiness (0–1), data are issued from European Centre for Medium-Range Weather Forecasts (ECMWF) fields (at the first vertical



**Figure 2.** Overview of meteorological and chemical conditions occurring during the summers of (left) 1998 and (right) 1999.

level, i.e.,  $z \approx 35$  m) and are diurnally averaged over a day. Figure 2 (bottom) corresponds to AIRPARIF network  $[\text{O}_3]$  ( $\mu\text{g m}^{-3}$ ) surface measurements: For each day the value displayed corresponds to the  $[\text{O}_3]$  peak over the whole Paris area. Figure 2 indicates that the mean wind speed varied from 0 to  $8\text{ m s}^{-1}$ . However, IOPs were always scheduled for relatively stagnant periods when wind speeds were  $< 2\text{ m s}^{-1}$

(except for IOP 4, where winds were  $3\text{ m s}^{-1}$ ). In 1998 and 1999, all IOPs occurred when temperatures were highest. For example, during IOP 2 the highest temperatures during the entire summer of 1998 were observed. In addition, cloudiness (integrated over the whole atmospheric column) was highly variable. During IOPs, averaged cloudiness was always  $< 0.4$  (with a maximum of 1). Finally, IOPs occurred

**Table 1.** Overview of the 12 ESQUIF IOPs<sup>a</sup>

IOP	Dates	Air Mass Origin	Remarks (Whole IOP)
<i>1998 Summer</i>			
1	25–26 July 1998	continental; 1–2 days stagnation	mostly cloudy by medium and high clouds; no wind
2	6–12 Aug 1998	continental; 2–4 days stagnation	clear sky; hot temperature; light winds
<i>1999 Summer</i>			
3	16–17 June 1999	continental; 3–4 days stagnation	overcast after 1200 UT; wind (NE) < 3 m s <sup>-1</sup>
4	25–26 June 1999	continental; 3–4 days stagnation	clear sky; wind (E) ~5 m s <sup>-1</sup>
5	1–2 July 1999	maritime	clear sky; wind (SE) ~4 m s <sup>-1</sup>
6	16–18 July 1999	maritime then stagnant	clear sky
7	24–25 July 1999	continental; 2–3 days stagnation	Paris plume tracking in Brittany; windy
8	28–31 July 1999	continental; 3–4 days stagnation	light wind <2 m s <sup>-1</sup>
<i>1999–2000 Winter</i>			
9	7–8 Oct 1999	continental; 2–3 days stagnation	anticyclonic situation; NO → NO <sub>2</sub> experiment
10	26–28 Jan 2000	continental	high [NO <sub>x</sub> ]; plume to west of Paris
<i>2000 Summer</i>			
11	19–20 July 2000	no photooxidant peak; moderate wind	aerosols experiments
12	30–31 July 2000	dry conditions; no stagnation	aerosols experiments

<sup>a</sup>The air mass is specified in order to check its continental or maritime origin. Additional remarks are displayed on the mean meteorological conditions observed during each IOP.

when highest [O<sub>3</sub>] concentrations prevailed, consistent with the situation for temperature.

## 2.2. Comparisons Between IOPs

[10] For the entire set of summertime IOPs, some meteorological values were similar, but different phenomena were also observed. Although the result was always an observed peak in oxidant levels, the meteorological situations were not identical. The differences were mainly due to differences in the synoptic origins of the air masses. A large portion of France lies along the coast of the Atlantic Ocean (west), whereas another part is surrounded by very industrialized countries (north and east). Thus relatively clean or relatively polluted air masses are advected over Paris, depending on the direction of the prevailing winds.

[11] For most of the IOPs, peaks in pollutants were observed when air masses remained for several days over large source regions such as Benelux and Germany to the north and the northeast of the Paris area (IOPs 1, 2, 3, 7, and 8). However, there were also occasions when high levels of pollutants were observed in conditions where air masses originated from the Atlantic Sea (IOPs 2, 5, and 6) (e.g., relatively clean maritime air). A common feature of these latter situations was the very weak winds. Consequently, even though air masses originated from over the sea during these IOPs, stagnant conditions occurred over Paris, accompanied by high temperatures and clear skies.

[12] On the basis of results from the IOPs, pollutants observed during the ESQUIF project can be split into two components: pollutants from air masses advected over the region and pollutants from local sources. Such a characterization of sources is a well-known problem in defining the chemical boundary conditions for initialization of models (see section 4.1). Thus another goal of the ESQUIF project was to quantify the transport of pollution from Paris to other regions. In the area surrounding Paris, such pollution consists of transport of “ozone plumes” originating in Paris. The impact of such plumes was quantified in terms of chemical regimes during IOPs 6 and 8, the results of which are presented by *Sillman et al.* [2003]. At still larger spatial scales, long-range transport of pollution several hundred kilometers

from the city (IOPs 4 and 7) is important. During ESQUIF this issue was studied using the technique of adjoint modeling sensitivity (described by *Schmidt and Martin* [2003]).

## 2.3. Comparisons Within IOPs

[13] Not only were there differences between IOPs, but meteorological and chemical variations were observed within an individual IOP. For example, during IOP 2, when the first day was characterized mainly by local production of oxidants, the end of the period was characterized by mixing of local sources with those transported long range [see also *Menut et al.*, 2000]. Whereas the beginning of the IOP (7 August 1998) brings mainly oceanic air masses (from the Atlantic Ocean) with low concentrations of pollutants, a change of the wind direction occurred during the IOP, and winds show that the air masses stagnated over strong source areas (Great Britain, the Benelux countries, and the Ruhr area in Germany). The observed local changes are, of course, influenced by synoptic changes. The impact of this is seen, in particular, in the vertical temperature soundings. From 7 to 9 August 1998 the height of the temperature inversion increases from 800 m (at the maximum) to 2800 m [*Hourdin et al.*, 2002]. These changes influence considerably the capacity of the boundary layer to vertically mix the primary pollutants emitted within the surface layer as well as the ability to exchange polluted air with relatively cleaner air from more distant regions at higher altitudes. A study based on lidar profiles performed in Palaiseau (25 km southwest of Paris) characterizes the coupling between the residual layer and the convective layer at the beginning of the day [*Fochesatto et al.*, 2001]. By comparison with simulations performed with the mesoscale MESO-NH model [*Lafore et al.*, 1998], it was shown that the residual layer could strongly interact and exchange with the convective layer and thus was not entirely isolated.

## 3. Pollutant Concentrations During the Summers of 1998 and 1999

[14] A synthesis of the average and maxima of surface ozone concentrations (in  $\mu\text{g m}^{-3}$ ) observed in Paris and in



**Table 2.** AIRPARIF Network Surface Station  $[\text{O}_3]$  Values Obtained During the Whole Set of the Summertime ESQUIF IOPs<sup>a</sup>

Date	Paris			Paris Area ( $150 \times 150$ km)			
	$[\text{O}_3]$ , $\mu\text{g m}^{-3}$	$[\text{O}_3]_{\text{max}}$ , $\mu\text{g m}^{-3}$	Time, UTC	$[\text{O}_3]$ , $\mu\text{g m}^{-3}$	$[\text{O}_3]_{\text{max}}$ , $\mu\text{g m}^{-3}$	Site <sup>b</sup>	Time, UTC
<i>IOP 1</i>							
25 July 1998	61.4	115.0	15	64.6	146.0	SW	16
26 July 1998	61.4	135.0	15	64.7	148.0	S	14
<i>IOP 2</i>							
6 Aug 1998	45.3	147.0	15	52.8	157.0	NE	16
7 Aug 1998	41.0	149.0	13	54.9	<b>260.0</b>	SW	16
8 Aug 1998	57.5	172.0	11	71.9	<b>237.0</b>	W	15
9 Aug 1998	99.4	<b>218.0</b>	16	102.1	<b>244.0</b>	SW	16
10 Aug 1998	71.5	<b>226.0</b>	12	78.9	<b>225.0</b>	NE	12
11 Aug 1998	100.9	<b>274.0</b>	14	112.0	<b>340.0</b>	NE	15
12 Aug 1998	69.6	134.0	2	75.8	129.0	W	2
<i>IOP 3</i>							
16 June 1999	54.2	150.0	13	65.7	<b>211.0</b>	SW	14
17 June 1999	56.9	133.0	12	73.0	176.0	SE	13
18 June 1999	49.7	97.0	16	58.4	115.0	SW	15
<i>IOP 4</i>							
25 June 1999	78.4	145.0	13	86.7	167.0	SW	16
26 June 1999	61.0	117.0	12	66.1	138.0	NE	12
<i>IOP 5</i>							
1 July 1999	27.1	68.0	12	33.7	94.0	SE	10
2 July 1999	54.8	119.0	13	58.0	<b>186.0</b>	NW	14
3 July 1999	43.9	81.0	10	47.5	101.0	NE	11
<i>IOP 6</i>							
16 July 1999	64.2	130.0	16	64.7	163.0	E	17
17 July 1999	74.3	166.0	12	78.8	<b>266.0</b>	W	16
18 July 1999	95.3	179.0	13	88.3	<b>191.0</b>	NW	12
19 July 1999	68.9	114.0	15	66.0	123.0	SW	14
<i>IOP 7</i>							
24 July 1999	51.2	99.0	15	60.3	126.0	SW	14
25 July 1999	67.8	118.0	23	70.2	132.0	W	22
26 July 1999	72.8	135.0	16	83.4	163.0	SW	15
<i>IOP 8</i>							
28 July 1999	76.1	140.0	13	93.4	<b>185.0</b>	SW	15
29 July 1999	78.5	165.0	15	94.9	<b>228.0</b>	SW	15
30 July 1999	82.8	<b>207.0</b>	14	100.0	<b>242.0</b>	W	15
31 July 1999	72.1	169.0	11	92.8	<b>206.0</b>	NW	13
1 Aug 1999	66.3	156.0	15	88.4	<b>189.0</b>	W	15

<sup>a</sup>Paris values display values within the city. Paris area corresponds to a domain covering  $150 \times 150$  km centered on Paris city. Concentrations exceeding the “public alert” threshold of  $180 \mu\text{g m}^{-3}$  are shown in boldface.

<sup>b</sup>Position of the surface station relative to Paris.

the entire Paris area are shown in Table 2. The hours corresponding to these peaks are also reported. For the “regional” peaks, also reported are the positions of the stations relative to Paris (for example, W represents a station in the west of Paris). Concentrations exceeding the “public alert” threshold of  $180 \mu\text{g m}^{-3}$  are shown in boldface.

[15] The concentrations of ozone within Paris are always less than those over the whole region. This is explained by the titration of ozone by NO, which is most strongly emitted within the city. For the few days when this is not the case it is noted that the “regional” peak is, in fact, observed for a peri-urban station very near the city. This feature highlights the problem of representativity of ground sampling. The characterization of the representativeness of surface stations for analysis and data assimilation is of primary interest and constituted one of the main research thrusts of this project [Blond, 2002].

[16] Within the city of Paris, peaks in ozone were observed 1–3 hours before the maxima over the entire region were observed. This is explained by the transport of the ozone plume. Under weak wind conditions the plume remains within the region of the city, and its concentration continues to increase as it travels over the strong peri-urban sources. The majority of the peaks occur in midafternoon during maximum solar insolation. They occur during hours that vary widely between 1100 and 1600 UTC. This range is the result of small-scale meteorological processes that are poorly sampled and thus badly simulated. In particular, the peaks are determined by the urban atmospheric boundary layer (ABL) growth rate and maximum depth and by the role of the entrainment zone as well as by the role of thin residual layers where ozone is trapped. One notable exception to this is the peak that formed on 12 August 1998 (IOP 2). In this particular case a major pollution episode is

ending, and the strong nocturnal concentrations observed correspond to ozone produced during the previous day (11 August, the most polluted day of summer 1998).

#### 4. Main Research Topics

[17] The ESQUIF experiment was designed to provide a more complete description of the atmospheric chemical composition above and around Paris city. This goal was achieved by the use of numerous aircraft measurements at several ranges from the city center and at different altitudes. The field studies were devoted, in part, to the examination of the chemical boundaries of the studied domain. This was the first step in the characterization of the relative magnitudes of advected air masses on local pollutants. In the region surrounding Paris, studies were carried out to evaluate the impact of photolysis on chemical reaction rates. Within the Paris area, comparisons were made between the large set of measurements and model simulations constrained by local emissions data [Vautard *et al.*, 2003].

[18] Another part of the field research component was devoted to the small-scale processes occurring within the studied domain. This mainly concerns the vertical exchanges of pollutants within the boundary layer, the stability of the residual layer [Fochessatto *et al.*, 2001], and a new nonlocal mixing parameterization in the dynamical Global Climate Model of the Laboratoire de Meteorologie Dynamique (LMDz) [Hourdin *et al.*, 2002].

##### 4.1. Regional- and Continental-Scale Interactions

[19] With no important orographical forcings, over the Paris area, one can expect a fairly simple meteorology that is relatively easy to simulate. However, in the absence of dominant forcing it is also necessary to better understand meteorology at longer distances. Even if the main goal of the project remains the Paris area ( $\sim 100 \times 100$  km), from the very start of the project it appeared that it was necessary to carry out measurements over the whole of northern France ( $\sim 800 \times 800$  km). This justified the choice of aircraft measurements at the mesoscale to understand the origins of the air masses entering Paris. In the same way, this explains the IOP 7 measurement strategy, initiated in order to follow the Paris ozone plume toward the west of France.

[20] The experimental results highlight the limitations of CTMs that employ “climatological” boundary conditions. Although studies of other polluted cities (those with local dominant forcings only) that employ “climatological” boundary conditions have been successful, this approach is not possible for the Paris region. Consequently, two new CTMs were developed, named MOCAGE [Peuch *et al.*, 1999] and CHIMERE [Vautard *et al.*, 2001; Schmidt and Martin, 2003], from the regional to the global scale.

[21] Determination of accurate boundary conditions requires the use of a realistic model at a larger scale than the studied area. The Paris area may appear as a “very urbanized island” in the middle of great rural areas. Thus, to have realistic simulations of pollutants in these rural areas, the large-scale CTM needs realistic biogenic emissions. The ESQUIF project represented a good opportunity to more precisely study these biogenic emissions [Sarrat, 2002; Derognat *et al.*, 2003]. In the work of Derognat *et al.*

[2003] a biogenic VOC emissions database was developed and used with the CHIMERE model (continental and regional scales). This database was validated during ESQUIF by comparison between model simulations and aircraft measurements of isoprene. Results showed that the difference was  $<10\%$ . It was also shown that the continental contribution of biogenic VOCs to the Paris ozone plume may reach up to 35 ppb of ozone for temperatures  $>35^\circ\text{C}$  (for example, during IOP 2).

##### 4.2. Surface Emissions

[22] Because emissions are some of the key inputs of CTMs, much work during ESQUIF focused on this aspect of air quality modeling. Since 1998, the Paris area has had one of the best emissions inventories in France. This emissions inventory, provided by AIRPARIF, was used by all the model teams of the ESQUIF project. A critical comparison between surface and aircraft measurements and modeled concentrations of numerous VOCs is proposed by Vautard *et al.* [2003].

###### 4.2.1. Model Versus Measurements

[23] The study by Vautard *et al.* [2003] examines the accuracy of the emissions inventory at the regional scale. Starting from a reference run (calculation of OH fields), a modified chemical mechanism of type VOC + OH allows for the calculation of the concentration of individual VOCs. Qualitatively, and by comparison with aircraft measurements, the model reproduced well the spatial distributions of these emissions. However, it was shown that the model overestimates and underestimates various individual VOC species, whereas some other emitted species, such as  $\text{NO}_x$  and CO, were accurately reproduced in the emissions inventory.

###### 4.2.2. Model Sensitivity and Uncertainty

[24] For ESQUIF a Bayesian Monte Carlo uncertainty analysis was developed. Even if the measurement constraint from circular flights and ground-based sites allows significant reductions in the model uncertainty, it is still substantial, i.e., between 15 and 30% in ozone maxima, both for a reference and a 50%-reduced emission scenario [Beekmann and Derognat, 2003]. Sillman *et al.* [2003] characterized the sensitivity of the Parisian plume to  $\text{NO}_x$  and VOC concentrations and showed that the Paris area is mainly VOC-limited. Concerning emissions, and, more generally, the whole set of CTM parameters, Menut [2003] proposed a classification of the meteorological and chemical processes that influence the final simulated concentrations of  $\text{O}_3$ ,  $\text{O}_x$ , and  $\text{NO}_x$  (IOP 2, using an adjoint model).

##### 4.3. Photolysis

[25] Mainly produced by photolysis processes, ozone is particularly sensitive to the phenomena that attenuate solar UV radiation. This motivates two particular studies, the first one a direct comparison of measurements of  $J(\text{NO}_2)$  within Paris with the TUV model; the second, a study of chemical partitioning within aerosols. On the basis of these analyses (see R. Vautard and the ESQUIF Team, ESQUIF final report, 2001, <http://climserv.lmd.polytechnique.fr/esquif>) the values of  $J(\text{NO}_2)$  are only attenuated  $\sim 12\%$  compared with the modeled reference values. Moreover, it was found that the majority of particles had diameters not exceeding 300 nm. Another study, using the CHIMERE model,

allowed for implementation of a new parameterization of the attenuation of photolysis rates, one based on the use of ECMWF cloud cover. According to a statistical regression, and after simulation of the entire summers of 1998 and 1999, it was shown that explicitly accounting for cloudiness considerably improved the simulations of surface ozone concentrations over western Europe. This improvement is most significant in northern Europe, the area that is most cloudy.

#### 4.4. Data Assimilation

[26] In order to improve understanding of pollution processes, a major goal of this project was to improve the daily forecast of peak ozone abundances. Toward this goal a method based on optimal interpolation was developed for regional scales [Blond, 2002]. This approach, which mathematically combines model simulations and measurements, allows for the generation of maps of ozone concentrations that are as realistic as possible. This technique is now employed daily under the auspices of the Project for Daily Ozone Forecast Over Western Europe (PIONEER) <http://euler.lmd.polytechnique.fr/pioneer/>), dedicated to daily forecasts over Europe.

#### 5. Conclusion

[27] The ESQUIF project documented numerous episodes of photooxidant pollution in the Paris area. The measurements and results have improved our understanding of the processes that generate pollution within the Paris area as well as the contributions of transport into and out of the Paris region to local and regional pollution. Many research topics were addressed with specific results for the Paris area, but there were some results that can improve our understanding of other polluted cities.

[28] From all the intensive observation periods it was found that pollution events were observed even when stagnant conditions appeared only very recently. The maximum thickness of the ABL varies much during all the episodes, and it was observed that oxidant peaks occurred for low or high ABL height. Moreover, these episodes appeared even in the cases of not-so-marked temperature inversion.

[29] Alone, activities in Paris and its suburbs can produce pollution peaks that are often moderate. The most significant peaks are the result of this local production, amplified by meteorological conditions that favor weak winds (and high temperatures) and that confine air masses for several days to regions with strong emissions. This finding highlights the importance of the transport of pollutants between countries in western Europe and, from an attribution point of view, the need for highly accurate chemical boundary conditions.

[30] Lastly, we note that the entirety of the ESQUIF database is accessible at <http://climserv.lmd.polytechnique.fr/esquif>. This includes the continental model CHIMERE, developed during this project, whose sources and documentation can be found at <http://euler.lmd.polytechnique.fr/chimere>.

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