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Notes des Activités Instrumentales

MEASUREMENTS AND MODELING OF ATMOSPHERIC POLLUTION OVER THE PARIS AREA: AN OVERVIEW OF THE ESQUIF PROJECT

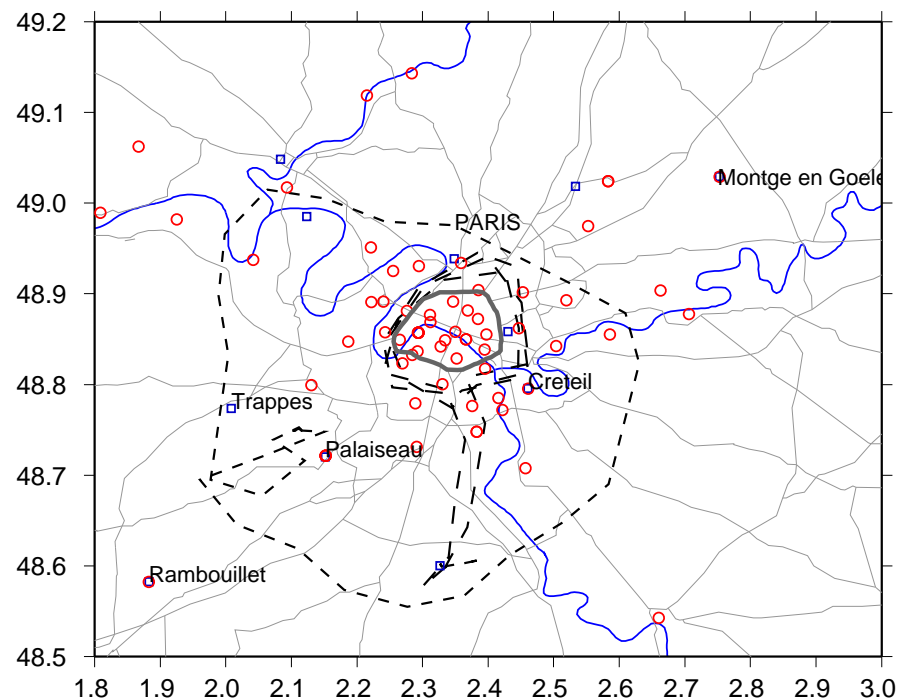
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Measurements and Modeling of Atmospheric Pollution over the Paris Area: An Overview of the ESQUIF Project

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Abstract. The "Étude et Simulation de la Qualité de l'air en Ile de France" (ESQUIF) project is the first integrated project dedicated to the study of the processes leading to air pollution events over the Paris area. The project was carried out over two years (summer 1998 to winter 2000) to document all types of meteorological conditions favourable to air quality degradation, and in particular to photo oxidant formation. The goals of ESQUIF are (i) to improve our understanding of the relevant chemical and dynamical processes and, in turn, improve their parameterizations in numerical models, and (ii) to improve and validate existing models dedicated to pollution analysis, scenarios and/or forecasting, by establishing a comprehensive and thorough database. In this paper, we present the rationale of the ESQUIF project and we describe the experimental set-up. We also report on the first experiments which took place during the summer of 1998 involving surface networks, remote sensing instruments as well as several aircraft. Focusing on three days of August 1998, the relative contributions of long-range transported and locally-produced ozone to the elevated ozone concentrations observed during this period are discussed and chemistry-transport model preliminary results on this period are compared to measurements.

Key words. Field experiment, urban pollution, ozone episode, aircraft measurements

1 Rationale and objectives of the ESQUIF project

As in many big cities throughout the world, pollution levels in the Paris area due to concentrated activity have become a major health issue. Current predictive capabilities must be

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improved to enable policy makers to reach balanced decisions on emission reduction strategies. This requires a thorough understanding of the processes and factors affecting the formation of pollutants and their concentrations.

Air quality over Paris is continuously surveyed by a dedicated surface network carrying out measurements of critical pollutant concentrations (AIRPARIF). However, surface measurements are not sufficient to fully understand the pollutants dynamics and chemistry. Also, unlike cities as Los Angeles (McElroy, 1993), Athens (Ziomas, 1998), Mexico-City (Doran *et al.*, 1998) where the local dynamical processes are dominant due to mountain barriers, and land-sea breeze regimes, the Paris agglomeration has a rather flat topography and is thus also strongly influenced by synoptic and local-scale meteorological conditions. Moreover, photochemical pollution in this area is actually both due to photooxidant build-up from local emissions and to large-scale transport of ozone (as studied over Boston during the North Atlantic Regional Experiment, Fast and Berkowitz, 1996) formed over other areas of Europe. The determination of their respective role, as well as several other scientific issues, requires a specific research project where numerical modeling and measurement campaigns are simultaneously performed. During the last decade, several campaigns have been carried out to study photochemical pollution at the regional scale. POLLUMET (Pollution and Meteorology) was carried out in Switzerland (1992-1994) and illustrated the influence of the Milan ozone plume (Prévôt *et al.*, 1997). BERLIOZ (Berlin Ozone) (Volz-Thomas *et al.*, 2000; Rappenglueck *et al.*, 2000) was designed to study the chemical reactions taking place in the plume of the city of Berlin (1998). LOOP-PIPAPPO (Limitation of oxidants production) focussed on chemical regimes in the Po valley, over Milan (Neftel, 1999).

ESQUIF ("Étude et Simulation de la Qualité de l'air en Ile de France", a French acronym for "Air quality study and modeling over the Paris area") is such a research initiative.

It is the first important project dedicated to the study of pollution in this area. The particular interest of the Paris area stems from its geographical situation: It is a large city located far from other populated areas by about 200 km. It is therefore in principle easier than for other European cities to distinguish the air masses charged with locally-emitted pollutants from those charged with pollutants emitted in remote areas. This should allow one to distinguish between errors coming from boundary conditions and from model parameterizations in regional model simulations. Another geographical interest of the Paris area is its low topography, which in the absence of local forcing makes the modeling of the accumulation/dispersion of pollutants more complex (Lossec, 1992; Dupont *et al.*, 1999). More specifically, the ESQUIF project focuses on several research issues, the most important ones listed here:

- The thermodynamics of the boundary layer in and around a big city: The main questions are (a) the role of the vertical structure of the boundary layer and the turbulent mixing processes during pollution episodes, and (b) whether models are able to simulate the air flow, its vertical structure and the surface fluxes encountered during low dispersion episodes, and, if not, how to improve the various parameterizations used within the models,
- The photochemistry of pollutants: the focus here is essentially on processes modifying the radiative fluxes and thereby photolysis rates (clouds, aerosols, total ozone column),
- The transport of pollutants at the interface of the urban/regional and the continental scale: we will evaluate the concentration of ozone and its precursors both at the inflow boundaries of the domain and in the city plume,
- The physical and chemical evolution of urban and sub-urban aerosols and their representation in air quality chemistry-transport models.

These research issues aim at giving some answers for the following environmental issues: (i) The origin of ozone episodes in the Paris area, including the determination of the relative role of local emissions and large-scale transport, (ii) The influence of the city area on neighbouring regions, including the study of the pollutants in the plume, their aging and the horizontal and vertical extent of this plume, (iii) The determination of the photochemical regimes (NO_x or VOC limited) and their spatial distribution, and (iv) The evaluation of existing emission inventories using measurements acquired during the campaign.

The project has been initiated jointly by the Institut Pierre Simon Laplace (IPSL) and the Laboratoire Inter-universitaire des Systèmes Atmosphériques (LISA), and the partnership is extended to the Laboratoire d'Aérodynamique, the Forschung Zentrum of Jülich, AIRPARIF (the local air-quality monitoring network) and Météo-France (see Table 1). The project extended over two years (July 1998 - January 2000).

The measurement campaign started during the summer of 1998, and ending during the winter of 1999-2000: Ten episodes of 1-5 days were documented, denoted hereafter the ESQUIF IOPs, (Intensive Observation Periods). These IOP days were selected on the basis of weather forecasts favourable to degradation of air quality. Table 2 lists the main

Table 2. Overview of the ten IOPs of ESQUIF from 1998 to 2000. The 'airmass' is specified in order to check its continental or maritime origin (with 'st.' for stagnation). T_{surf} and O_3 correspond to maximal surface temperature and ozone concentrations, respectively, observed over the Paris city during the diurnal day. Some additional remarks are displayed on the mean meteorological conditions observed over each whole IOP.

IOP	Dates	Airmass	Remarks (whole IOP)
1	25/07/1998 26/07/1998	Continental 1-2 days st. stagnation	Mostly cloudy by medium and high clouds no wind
2	6/08/1998 7/08/1998 8/08/1998 9/08/1998 10/08/1998 11/08/1998 12/08/1998	Continental 2-4 days st.	Clear sky Hot temperature Light winds
3	16/06/1999 17/06/1999	Continental 3-4 days st.	Overcast after 12UT. Wind (NE) < 3 m/s
4	25/06/1999 26/06/1999	Continental 3-4 days st.	Clear sky Wind (E) \approx 5 m/s
5	1/07/1999 2/07/1999	Maritime	Clear sky Wind (SE) \approx 4 m/s
6	16/07/1999 17/07/1999 18/07/1999	Maritime then stagnant	Clear sky
7	24/07/1999 25/07/1999	Continental 2-3 days st.	Paris plume tracking in Brittany. Windy
8	28/07/1999 29/07/1999 30/07/1999 31/07/1999	Continental 3-4 days st.	Light wind < 2 m/s
9	07/10/1999 08/10/1999	Continental 2-3 days st.	Anticyclonic situation $\text{NO} \rightarrow \text{NO}_2$ experiment
10	26/01/2000 27/01/2000 28/01/2000	Continental	High NO_x concentrations Plume to West of Paris

characteristics of the meteorological situations encountered during these IOPs.

Following the ESQUIF objectives, the campaign included aircraft and ground-based measurements. Various flight plans were defined: flights to document spatial ozone distribution in loops around the city at various distances from the center, the "city plume" flights to follow the urban plume, the "chemical regimes" flights occurring at constant latitude over the Paris area, and the "regular" flights performed to obtain a climatology of long-lived trace gas species around the Paris area.

The paper is organized as follows: in Section 2, we present the main defined experiments and the experimental strategy designed to address the scientific goals in the framework of ESQUIF. The instrumental set-up is also briefly discussed. In Section 3, the models to be used in the framework of ESQUIF are briefly described. Section 4 is devoted to the description of the meteorological conditions and experimental set-up during the two IOPs performed during the summer of 1998 and to report on results obtained during the second IOP. Chemistry-transport model results are presented and compared to measurements in Section 5. In Section 6 we discuss the perspectives of further work within the ESQUIF project.

Table 1. List of institutes and people involved in the ESQUIF experiment.

Institute, acronym and affiliation	Peoples involved in ESQUIF (in alphabetical order)
AIRPARIF (Parisian pollution network)	E.Gilibert, D.Gombert
Laboratoire Inter-Universitaire des Systèmes Atmosphériques (LISA) - Univ. Paris XIII IPSL/CNRS	S.Alfaro, B.Aumont, G.Bergametti, C.Boissard, F.Brocheton, B.Chatenet, A.Dutot, A.Gaudichet, L. Gomes, C.Jambert, M.Maille, T.Marion, F.Millard, P.Perros, G.Toupance
Laboratoire des Sciences du Climat et de l'Environnement (LSCE) - Commissariat à l'énergie atomique/IPSL, Saclay	B.Bonsang, P.Bousquet, H.Cachier, P.Chamaret, P.Chazette, P.Ciais, A. Foucras, D.Martin, P. Moral, C.Morfopoulos, D.Picard, M.Ramonet, R. Sarda, M.Touaty, P. Vaillant, C. Valant
Laboratoire de Météorologie Dynamique (LMD) - IPSL/CNRS -Paris & Palaiseau	H.Chepfer, P.Delville, P.Drobinski, P.H.Flamant, C.Honoré, F.Hourdin, C.Loth, L.Menut, J.Roux, R.Vautard
Météo-France (French Weather Service)	C.Abonnel, H.Bellec, D.Bernard, R.Caillou, M.Charpentier, G.Duverneuil, F.Goyon, J.C.Etienne, M.Laurens, M.P.Lefebvre, B.Lossec, M.Morera, M.Payen, V.H. Peuch, F.Pouvesle
Service d'Aéronomie (SA) IPSL/CNRS - Paris	O.Antivilo, G.Ancellet, M.Beekmann, C.Flamant, P.Genau, G.Mégie, J.Pelon, M.Pinharanda, J.P.Pommereau
Laboratoire d'Aérodynamique CNRS - Toulouse	D.Guédalia, R.Rosset
Forschungszentrum Jülich, D-52425 Jülich, Germany	D.Kley and METAIR

2 Overview of the experimental strategy and set-up

2.1 IOP organization and forecast

In order to trigger IOPs, several models and model outputs were used (see description below). First, we used weather forecasts issued at Météo-France and compared them to the European Centre for Medium-Range Weather Forecasts (ECMWF) and National Centers for Environmental Prediction (NCEP) forecasts. When favourable weather conditions were forecasted and when the forecast became stable (typically 2-3 days before the experiment), we checked the chemistry-transport forecast model CHIMERE (Menut *et al.*, 2000). This model is forced either by ECMWF or Météo-France meteorological variables (wind, temperature, humidity, cloudiness) and forecasts pollutant fields (ozone in particular) which were used to trigger an IOP. A particular emphasis was put on the nature of the episode (local-scale or large-scale). The availability of airplanes and instruments was also taken into account in the design of the experiment (a complete list of instruments is displayed in the Table 3). Finally, the IOPs were chosen to be meteorologically as different as possible in order to collect a large range of situations (e.g. continental air, maritime air, hot weather, stagnation conditions).

2.2 Aircraft measurements

Five aircraft (two Piper AZTECs, a MERLIN IV, the Fokker 27 "ARAT" and a DIMONA) were used to loop around Paris at various distances and to collect measurements of CO, NO, NO₂, O₃ and NMVOC's (Non-Methane Volatile Organic Compounds) as well as standard meteorological parameters. The flight plans may be defined as (1) 'local and regional' flights (Figure 1), (2) 'meso-scale' flights (Figure 2), (3) 'dedicated' flights (urban plume and chemical regimes) and (4) the 'regular' flights, devoted to the CO₂ climatology, as follows:

2.2.1 The 'local and regional' flights In order to study the local and regional pollutants concentrations around the Paris agglomeration, three majors flight patterns were designed as loops around the city (Figure 1):

- 'Urban' flights, made above the Paris "ring" (limit of the Paris city) at 500, 1000 and 1500 m AGL, aim at characterizing pollutant concentration above the main sources (traffic, industry).
- 'Suburban' flights, made at a distance of 20 km from the center of Paris, were designed to document the chemical compounds concentrations at the border of the urban area. In addition, these flights will also be used for determining the gradient of concentrations between upwind and downwind the city, in order to help evaluating emission inventories.
- The 'Ile de France' flights (from now called 'IdF' flights), made at a distance of 50 km from the center of Paris, were also designed to explore the vertical distribution of pollutants by means of soundings at 4 points in each direction from the city, and to characterize pollution at rural sites close to the city.

2.2.2 The 'meso-scale' flights The 'meso-scale' flights (from herein called 'Meso' flights) were designed to document pollutant concentration fields (ozone, in particular) at a distance of 150 km from the center of Paris (Figure 2). These flights were performed with the MERLIN aircraft (Table 4) and the flights were done from one to two times per IOP (depending on the meteorological situation). These 'Meso' flights, along with the 'IdF' flights, are to be used to provide boundary conditions for regional air quality models, and to understand the transport of pollutants at the regional scale.

2.2.3 The 'dedicated' flights The 'dedicated' flights were designed to provide a database on the whole Paris area (130 x 120 km). These data include ozone, nitrogen compounds (NO, NO₂, NO_y, HNO₃, PAN) and NMHCs (C₂ - C₄) measured by the ARAT aircraft (Table 4) in the boundary layer using a regular trajectory (5 legs of 130 km separated by

Table 3. Description of the ground measurements deployed during the ESQUIF experiments. P , T , RH , U , V , q and z designate pressure, temperature, relative humidity, zonal and meridian components of the wind, specific humidity and altitude, respectively. O_3 , CO , etc... refer to concentrations for ozone, carbon monoxide, etc... The (*) and (+) symbols referred to the instruments in the LSCE “MAS” (for Mobile Aerosol Station) and LISA “MILEAGE” mobile stations, respectively.

Measurements	Instruments	Site	Measurements
Dynamics	Lidar	Palaiseau	ABL depth, entrainment zone depth
	Soundings	Trappes	P_z , T_z , RH_z , U_z , V_z , q_z
	Surface network	IdF	P_{2m} , T_{2m} , RH_{2m} , U_{10m} , V_{10m}
	Sensors*	Paris, Saclay	T_{2m} , RH_{2m}
	Station	Créteil	T_{20m} , RH_{20m} , u_{20m} , v_{20m}
Aerosol	Lidar	Palaiseau	Aerosol optical depth, extinction coefficient
	Lidar	Paris	Aerosol optical depth, extinction coefficient
	Nephelometer*	Paris, Saclay	Aerosol scattering coefficient
	Aethalometer*	Paris, Saclay	Surface black carbon concentration
	Aerosol sampling line*	Paris, Saclay	Chemical apportionment
	Particule counter*	Paris, Saclay	Aerosol size distribution (\varnothing : 0 \rightarrow 3 μ m)
	Diffusion battery*	Paris, Saclay	Aerosol size distribution (\varnothing : 7nm \rightarrow 0.2 μ m)
	TEOM balance*	Paris, Saclay	PM1 or PM10 Aerosol mass concentration
	J(NO ₂)	Paris	NO ₂ photolysis frequencies
	TEOM balance	Créteil	PM1 or PM10 Aerosol mass concentration
Cascad impactor	Créteil	Aerosol mass size distribution (0.03 to 10 μ m)	
Chemistry	SANOA	Paris	O ₃ , NO ₂ , NO, SO ₂ , VOC's
	SAOZ	Verrieres	O ₃
	Surface network	IdF	O ₃ , NO ₂ , NO, SO ₂ , CO, PM ₁₀
	Photometers	Paris, Palaiseau Saclay, Créteil	Optical thickness, Angström exponent size distribution
	Analyzers ⁺	Créteil	O ₃ , NO ₂ , NO, PAN, NMVOC's

30 km). The aim of the chemical regimes experiment is to build a map of the area surrounding Paris with respect to the NO_x and VOC sensitivity regimes under meteorological situations typical of high pollutant levels. This is done using indicator species (Milford *et al.*, 1994; Sillman, 1995) such as O₃/NO_y, O₃/NO_z and O₃/HNO₃. Some other ARAT flights were designed to study the urban plume: perpendicular flight legs to the mean wind direction were performed and gave pollutants gradients in and around the city plume.

In the same manner, chemical regimes will be studied using the DIMONA flights (Lehning *et al.*, 1998). The DIMONA aircraft (see Table 4 for measurements) circumnavigated the city of Paris, well inside the mixing layer, at a distance of about 30 km from the city center. Designed according to a trifold strategy, the first objective was to obtain measurements of the VOC/NO_x concentration ratios at a point close in time to the emissions. A short extrapolation to zero time will be made using the concept of chemical age (where the Chemical Age (CA) is the time integrated OH concentration from emission to the time of measurement, according to $\ln(\text{NO}_y)/(\text{NO}_x) = k(\text{OH})dt = k \text{CA}$, where k is the rate coefficient of oxidation of NO₂ to HNO₃). Here, the fact is used that emissions of the oxides of nitrogen are to a large extent in the form of NO. Photochemical equilibrium between NO and NO₂ is rapidly established within 1-2 minutes and the consecutive oxidation of NO₂ to HNO₃ by OH occurs with a known rate coefficient. Then CA can be derived from the measured ratio of NO_y to NO_x concentrations. The second objective was to experimentally determine integrated NO_x and VOC emission rates over the Paris area. This can be done, once the chemical age has been determined, from quasi-continuous NO_x and VOC measurements along the circumferential flight track. As an additional parameter the mix-

ing layer height must be known. The third objective centered around the concept of oxidant O_x (O₃+NO₂). From measurements of O_x on the circumference and using 3D wind field informations (measurements and model), it will be possible to derive the oxidant formation rate within the city and its export to the region.

2.2.4 The 'regular' flights Regional aircraft soundings in the vicinity of Orleans (48°N 1°E, 150 km South of Paris) have been carried out since April 1996, and since 1999 as part of the ESQUIF project. The objective of these flights is twofold: (1) to establish a climatology of long-lived trace gas species which are radiatively active (CO₂, CH₄) or intervene in the oxidizing capacity of the atmosphere (CO, Non Methane Hydrocarbons), and (2) to establish boundary conditions of continental “background” rural concentrations of CO and Non Methane Hydrocarbons (NMHCs) before their horizontal transport over the Paris Area. This is especially important in the case of NMHCs which intervene in the formation and destruction of ozone in polluted air, and can be emitted by vegetated areas outside of the urban area. The frequency of the regional flights was biweekly and the soundings were taken approximately at mid-day, between 50 m and 3000 m. The total flight time was on the order of 4 hours and the profile itself took about 1 hour. Pressure, air temperature and relative humidity were measured routinely along the profile. Pyrex glass flasks were sampled in pair at 7 different vertical levels (from 100 to 3000m) (Francey *et al.*, 1996, Gros *et al.*, 1998, Touaty, 1999).

The time series of CO vertical profiles over Orleans shows a well defined seasonal cycle within the free troposphere, with a maximum of 190 ppb in December-January and a

Table 4. Description of the flights measurements deployed during the ESQUIF experiments.

Compound & aircraft	Detection Technique	Detection Limit	Resolution Time (sec)
<i>ARAT INSU</i>			
O ₃	UV absorption	1 ppb	1
NO	Chimiluminescence	20 ppt	30
NO ₂	UV photolysis + Chimiluminescence	20 ppt	30
NO _y	Au-CO convertor + Chimiluminescence	20 ppt	60
HNO ₃	Nylon filter+ Au-CO convertor + Chimiluminescence	20 ppt	60
PAN	Gas Chromatography ECD	15 ppt	300
CO	Gaz IR filter correlation	50 ppt	30
NMVOC's	Onboard Gas chromatograph (C2 - C4)		
Black Carbon	Aethalometer	200 ng/m ³	15
<i>DIMONA</i>			
O ₃	UV absorption	1 ppb	1
NO	Converter + Chimiluminescence (Luminol)	0.5 ppb	10
NO ₂	Chimiluminescence (Luminol)	0.5 ppb	10
NO _y	Converter + Chimiluminescence (Luminol)	0.5 ppb	10
HNO ₃	Converter + Chimiluminescence (Luminol)	0.5 ppb	10
PAN	Converter + Chimiluminescence (Luminol)	0.5 ppb	10
CO	IR absorption		
NMVOC's	In situ Gas chromatography (C4 - C10)		
<i>MERLIN IV Météo-France</i>			
O ₃	UV absorption	2 ppb	7
NO	Chimiluminescence	100 ppt	30
NO _y	Mo convertor + Chimiluminescence	150 ppt	30
CO	Gaz IR filter correlation	50 ppt	30
<i>AZTEC Météo-France</i>			
O ₃	UV absorption	2 ppb	5
NO	Chimiluminescence	1 ppb	50
NO _y	Mo convertor + Chimiluminescence	1 ppb	50
CO	Glass Canisters + Gas chromatography		
NMVOC's	Glass Canisters + Gas chromatography (C2 - C7)		
CO ₂ , δ ¹³ C, δ ¹⁸ O	Gas chromatography + mass spectroscopy		
<i>AZTEC CEA-LSCE</i>			
O ₃	UV absorption	2 ppb	7
NO ₂	Luminol Chimiluminescence	100 ppt	1
CO	Gaz IR filter correlation	75 ppt	30
CO	inox flasks + Gas chromatography		
NMVOC's	inox flasks + Gas chromatography (C2 - C7)		
Black Carbon	Aethalometer	200 ng/m ³	15
CO ₂	pyrex flasks + IR absorption		
CH ₄	pyrex flasks + Gas chromatography		

minimum of 90 ppb in June-July-August. Free-tropospheric air is representative of Western European continental background air, where industrial activities, heating and transportation emit CO throughout the year while CO is destroyed by OH radicals during the summer. There is also a well defined seasonal cycle within the Atmospheric Boundary Layer (ABL), although the variability of local sources and of their mixing within the air column during the same day does not allow the sporadic profiles to define a real "climatology" of CO near the ground. Large accumulations of CO (up to 350 ppb) below 1000 m are discernable in winter. In summer the CO gradients between the surface and the troposphere is reduced, likely due to vigorous mixing and dilution of the surface sources into the upper layers.

2.2.5 Ozone analyzer comparisons Because ozone is the key compound of the campaign, different analyzers were used during the ESQUIF experiment. Thus, it was necessary to in-

tercompare and calibrate them in order to construct a homogeneous database. The intercomparison was performed during the watch period on July, 27 1999 (between IOP7 and IOP8). Two kinds of comparisons have been carried out. The first concerned the comparison in a laboratory (LSCE) and the second was an in-flight comparison. Laboratory comparisons were made between the 'Thermo Electron' ozone analyzers (ARAT aircraft, LISA and LSCE/CEA mobile stations, see Tables 3 and 4) and a Thermo Electron calibrator 49PS analyzer. It was found that the instruments had less than 5% calibration difference. The in-flight comparisons (MERLIN, Aztec LSCE and Aztec Météo-France) were performed on July 29, 1999, at two different altitudes (150m and 900m) when the aircraft flew on patrol. The detailed data are still being processed.

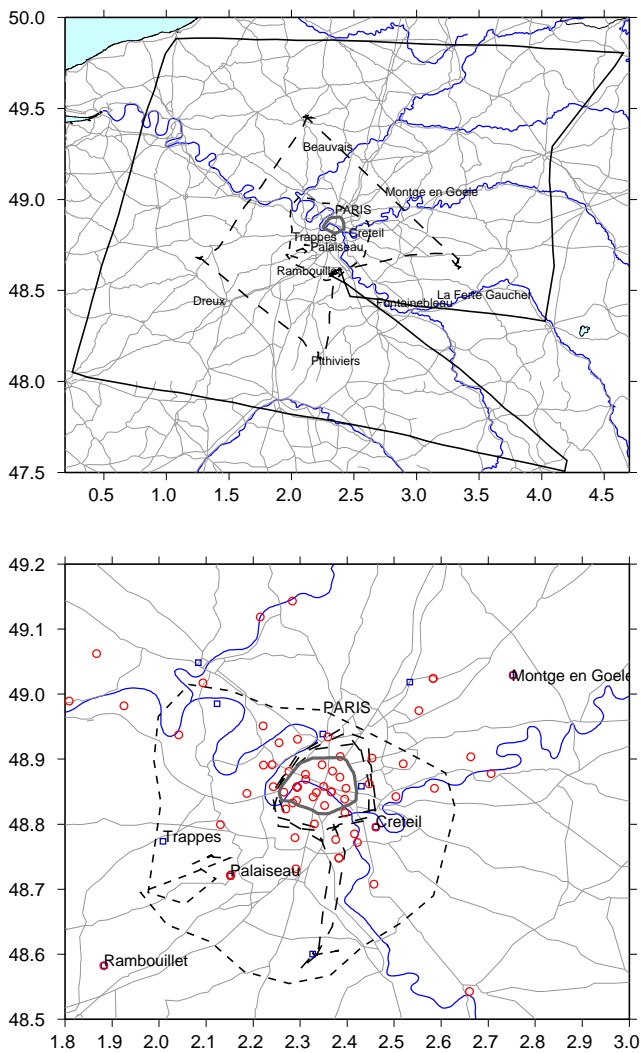


Fig. 1. The flights plans over the Paris area (the city of Paris is located at the center of the map). The symbols, 'squares' and 'circles', indicate the Météo-France and AIRPARIF surface stations (bottom), respectively. Typical flight patterns for 'Urban' (bottom), 'Suburban' (top and bottom), 'IdF' (top and bottom) and 'meso' (top) flights are given by the inner solid line, dotted line and dashed lines. The city names indicate locations of the main IOPs measurements on the regional Figure (bottom).

2.3 Ground-based measurements

2.3.1 Network measurements The AIRPARIF air quality network consists of more than 70 automatic stations performing continuous surface measurements of O_3 , NO, NO_2 , SO_2 , CO and PM_{10} concentrations (denoted as small circles in Figure 1 at three types of locations: urban-polluted, urban background and rural.). The Météo-France meteorological surface network performs hourly measurements of temperature, relative humidity and pressure (hPa) at 2 m above ground level (AGL), along with the wind speed and direction at 10 m AGL

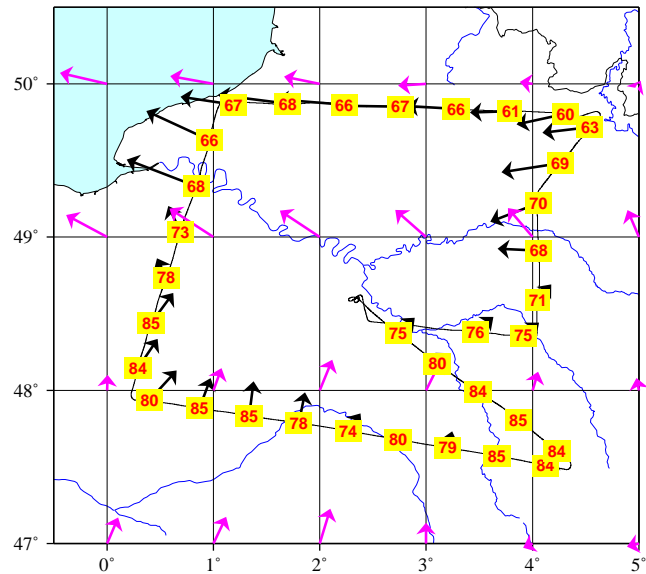


Fig. 2. A typical meso-scale flight pattern. As on the Figure 1, the city of Paris is at the center of the map. Values displayed on the Figure indicate ozone measurements (ppb) performed by the MERLIN aircraft on the 9A98 (0451-0826 UTC, 900m). Black vectors indicate the wind speed and direction along the flight trajectory, and gray vectors indicate the wind field modelled with the ARPEGE model.

(small squares on the Figure 1). As a first step, those measurements are used for model validation (see Section 5).

2.3.2 Chemical and aerosols measurements In addition to aircraft and air quality network measurements, several ground-based instruments were operated during the ESQUIF IOPs, and for some of them, during the whole summer.

In the center of Paris, high frequency (2Hz) measurements of O_3 , NO, NO_2 , SO_2 , HNO_3 and Volatile Organic Compounds (VOC's) concentrations were made routinely by a differential optical absorption spectrometer operating in the visible and the ultra-violet range (the "SANOVA" instrument, french acronym for 'Système d'ANalyse par Observation Active'). At the same location, measurements of NO_2 photolysis rates were performed from a JNO_2 radiometer. In addition, measurements were performed from the "Mobile Aerosol Station" instrumented van (MAS, in Table 3) in order to characterize the urban aerosols. The equipment included sampling lines for total aerosol and carbonaceous aerosol mass concentrations, a JNO_2 radiometer, an aethalometer, a nephelometer, a condensation nucleus counter and $PM_{2.5}$ and PM_{10} devices. This van was used at several places, depending on the experiment: for example, in the Paris city center during the IOP6 and in Saclay during the IOP7 (in conjunction with the aerosol measurements performed with the ARAT aircraft during these periods). During the entire summer of 1999, two multi-spectral photometers (Holben *et al.*, 1998) were used to measure aerosol spectral properties over Paris and Saclay. As aerosol optical properties are directly dependent on their age and primary sources, differences between the spectral dependency and the Angström exponent enable one to characterize

the aerosol regional transport. For example, during IOP7, in which Saclay was under the Paris plume, urban aerosols were compared with rural ones. Around noon, the urban optical thickness was twice as large as the rural one, while at the end of the day, optical thickness was comparable. For this case, we hypothesize that the urban aerosol, mainly generated by car traffic (black carbon and organic carbon), was advected over Saclay.

In the suburb of Paris, integrated ozone content was measured in Verrières (20 km, southwest of Paris) by a spectrometer operating in the visible and the ultra-violet (SAOZ, french acronym for “Système d’Analyse par Observation Zénithale”) twice a day, at dusk and dawn. This integrated ozone column is an important input parameter for an exact calculation of $J(\text{O}_3^{1D})$ photolysis frequencies with radiative transfer models. In addition, aerosols size distributions and composition (as well as their optical properties) were characterized in Saclay (25 km, southwest of Paris) using a nephelometer, an aethalometer and a condensation nuclei counter as well as total aerosol filters and black carbon filters.

2.3.3 Backscatter lidar measurements Two upward-looking backscatter lidars operating at a wavelength of $0.53 \mu\text{m}$ were deployed in the center of Paris and in Palaiseau (25 km southwest of Paris). The lidar was intended to document some of the key parameters relevant to (i) the ABL dynamics, i.e., the ABL depth and entrainment zone depth, (ii) radiative budget i.e., optical depth, and (iii) oxidation/production of photochemical species, i.e., extinction coefficient profiles (which are relevant to the actual photolysis rate), with a vertical resolution of 30 m. In addition, a pyranometer and a pyrgeometer (for measurements of infrared and longwave radiations), a sonic anemometer (turbulence), and pressure, humidity and temperature measurements were performed simultaneously to lidar measurements at Palaiseau. In addition, a differential absorption lidar (ALTO, acronym for “Airborne Lidar for Tropospheric Ozone”, Ancellet and Ravetta, 1998) was operated during the IOPs of summer 1999 at the Palaiseau site in order to continuously record the tropospheric ozone profile up to 5 km height.

2.3.4 Soundings The vertical thermodynamical structure of the atmosphere in the Paris area was documented by means of soundings performed by Météo-France in Trappes (25 km southwest of Paris). Temperature, relative humidity, pressure and wind speed and direction soundings were measured twice daily at 0000 and 1200 UTC. During some IOPs, additional soundings were performed every three hours at the Trappes site. From temperature, specific humidity and wind measurements, Richardson number and potential temperature profiles can be obtained which are then used to compute the diurnal evolution of the depth of the ABL. The ABL depth cycle can in turn be compared to that derived from lidar measurements.

3 Overview of numerical models

3.1 Meteorological models

Two meso-scale models, MERCURE (Souffland, 1985) and Meso-NH (Lafore *et al.*, 1998) will be used to perform simulations of the meteorological conditions on domains ranging from $100 \times 100 \text{ km}^2$ to $500 \times 500 \text{ km}^2$, centered on Paris. Horizontal and vertical resolutions may be adapted from 1 to 10 km and from 10 to 50 m, respectively. These models have already been used for simulating the air flow over Paris (Menut *et al.*, 1999a and Tulet *et al.*, 1999 for Mercure and Meso-NH, respectively). Realistic meteorological boundary conditions for these meso-scale models will be provided by the ECMWF and ARPEGE (Météo-France) analyses. In addition, a zoomed version of the Laboratoire de Météorologie Dynamique (LMD) general circulation model (LMDz, Hourdin and Armengaud, 1998) was also used (Hourdin *et al.*, 2000). The size of the grid cell characterizing Paris in this model is $50 \times 50 \text{ km}^2$. The grid size increases away from the center of the domain, and the model uses ECMWF for boundary conditions.

3.2 Chemistry-Transport models

The meteorological fields provided by the models described in the previous section will be used as inputs to the models presented here. At the local scale, the urban CHIMERE model (Vautard *et al.*, 1999; Menut *et al.*, 2000) was used as a forecast (Vautard *et al.*, 2000) and analysis tool. It is a more realistic adaptation of the box model approach proposed by Jin and Demerjian (1993) to the Paris area. It uses the MELCHIOR chemical mechanism of Beekmann and Lattuat (2000). At the meso-scale, the models used will be Meso-NHC (Tulet *et al.*, 1999), A₃UR (Jaeger-Voirol *et al.*, 1998) and POLLUTO, which are coupled to meteorological models Meso-NH, MERCURE and ECMWF data respectively. The chemical mechanism MoCA (Aumont *et al.*, 1997) of A₃UR has already been tested over Paris.

In addition, modified versions of CHIMERE, with an Eulerian grid, are currently being developed: namely a meso-scale version to perform simulations on a $150 \times 150 \text{ km}^2$ domain around Paris with a horizontal resolution from 6 to 10 km and a continental version over western Europe, with a $1/2$ degree resolution. Modeling results presented in this paper have been performed with the meso-scale chemistry transport model (CTM) POLLUTO. POLLUTO, is a three-dimensional chemistry-transport model using a cartesian mesh. The domain is defined in order to have the city of Paris (the most urbanized area) at the center of the horizontal mesh and is composed of $15 \times 9 \times 6$ nodes in the x-y-z directions, respectively. The model is forced by the ECMWF first guesses (6-hour forecasts). These meteorological fields are re-interpolated over the studied domain in time (one hour frequency) and space (with an horizontal resolution of 10 km and ranging from 0 to 2700 m vertically). To account for thermodynamic processes, POLLUTO requires mass fluxes, temperature, density, specific humidity and cloudiness (for chemistry) as inputs. At each grid point, turbulence parameterizations are used in order to retrieve vertical turbulent diffusiv-

Table 5. Overview of the meteorological conditions over Paris and maximum surface ozone concentrations in representative locations during the IOP 2. The background and plume concentrations are from the AIRPARIF stations in Montge en Goële and Rambouillet, respectively. The times were the maximum of ozone occurred are given in parentheses.

Day	7A98	8A98	9A98
<i>Meteorological conditions over Paris</i>			
Wind direction (from)	N-E	E	S to E
Wind speed at 10 m (m s^{-1})	1.4	1.3	1.8
Maximum ABL depth (m)	800	2300	2800
Temp. at 2 m ($^{\circ}\text{C}$) - low	16.5 (05)	17.5 (06)	18.7 (05)
Temp. at 2 m ($^{\circ}\text{C}$) - high	31.0 (16)	35.3 (16)	35.0 (15)
<i>Ozone concentrations (ppb) over the Ile de France (IdF)</i>			
Paris	60 (16)	85 (12) 55 (16)	95 (16)
Urban plume	125 (16)	90 (12) 85 (16)	120 (16)
Background	55 (15)	70 (12) 55 (16)	90 (16)
Production by Paris	70	20-30	30

ity profiles, surface friction velocity for dry deposition (Wesely, 1989). The gas-phase chemistry mechanism is MoCA and the adjoint part of the whole model is developed, and currently used for sensitivity analysis (see Menut *et al.*, 2000 for the adjoint principle). Hourly values of surfacic anthropic emissions are given for 15 primary pollutants, using two data sources: the CITEPA (1993) inventory, providing annual totals of emissions of NO_x , SO_2 , CO, CH_4 and NMVOCs (non speciated), and the GENEMIS (1994) data base, used for NMVOC speciation and temporal disaggregation of annual totals to hourly values.

The MOCAGE model (Peuch *et al.*, 1999), currently developed at Météo-France/CNRM, will be used to provide chemical boundary conditions at continental scale. Finally, as a complement to all previous Eulerian models, the Lagrangian model, MELCHIOR (Beekmann and Lattuat, 2000) was used for analysis of pollutant trajectories over Europe.

4 IOP2 of ESQUIF: Data analysis

The ESQUIF experiments performed during Summer 1998 have been designed to address more specifically the following questions: (i) what are the relative contributions of local production and long-range transport to the ozone concentrations measured over the Paris area during pollution events and (ii) what is the impact of the vertical boundary layer structure on ozone build-up over the Paris area? In this section, we perform preliminary analyses in order to illustrate how ESQUIF data can be used to address these questions.

In this paper, we focus on IOP 2 which lasted three days, from Friday to Sunday 7-9 August 1998 (hereafter called 7A98, 8A98 and 9A98, respectively, Table 5). This period (including 10 and 11 August 1998) corresponded to the highest ozone concentrations recorded during the whole summer of 1998. As the experimental set-up was optimum during

7A98, 8A98 and 9A98 (including aircraft measurements), we focus our analysis on these three days.

In order to understand the origin of air masses advected into the Paris area, a series of 7 day back-trajectories were calculated. These trajectories used the wind field as diagnosed by ECMWF first guesses. During the convective period, air parcels are well mixed within the boundary layer. Hence neither constant-altitude trajectories nor trajectories following the mean 3-dimensional wind field would be realistic. In order to overcome this problem, air trajectories were assumed to undergo a random altitude change every hour during the convective period (0900-1800 UTC). They were nevertheless bounded to stay between 0 and 1500m. Fifty back-trajectories are calculated in this way (due to the convective processes, an ending point does not correspond to the same origin. In order to taken into account these several origins, fifty trajectories are studied). Figure 3 represents these “back-plumes” ending in Paris during the three IOP days and the day prior. On 6A98 and 7A98, the air mass has a maritime origin, while on 9A98 it has travelled over the British Isles as well as Belgium, the Netherlands and Luxemburg. We thus expect that on day 9A98 a significant part of ozone measured over Paris originated from precursors emitted over Northern Europe, while on day 7A98, most of the pollution was locally produced.

4.1 Regional ozone formation and continental transport

In order to quantify the amount of ozone produced in large cities such as Paris, one cannot use measurements taken within the Paris city limits since ozone formation is usually maximum in the city plume, about 50 km downstream of the Paris city (see for instance Tulet *et al.*, 1999).

On 7A98, the urban plume was observed southwest of Paris from aircraft and surface measurements. In this region, ozone concentrations measured by the MERLIN aircraft (Table 6) at about 500 m AGL reached about 110 ppb at the end of the afternoon, while those encountered in the northeast side of the city were about 60-70 ppb. Therefore, photochemical ozone production from local emissions was at least about 50 ppb during this day. The aircraft looped 6 times around the city during the afternoon on the same trajectory. Thus, one can plot, at each trajectory point, a time series of the ozone concentrations.

We represent in Figure 4 this time evolution for four selected points. The first (second) location, in the northeast (southwest) of Paris, is used to characterize the ozone concentration upstream (downstream) of Paris. The other two locations (southeast and northwest of Paris) are chosen such as to document ozone concentrations in the cross-wind direction. These locations are further referred to as NE, SW, SE and NW (Figure 4). On the lee side, downwind of Paris, the ozone increase is very pronounced while on the upwind side concentrations do not vary much during the afternoon. At the two other locations, concentrations reach intermediate values, with a peak occurring earlier than in the plume. The same behaviour is observed from ground stations. At Montgé en Goële (about 40 km NE of Paris), peak concentrations reach about 60 ppb while at Rambouillet (southwest of Paris), peak

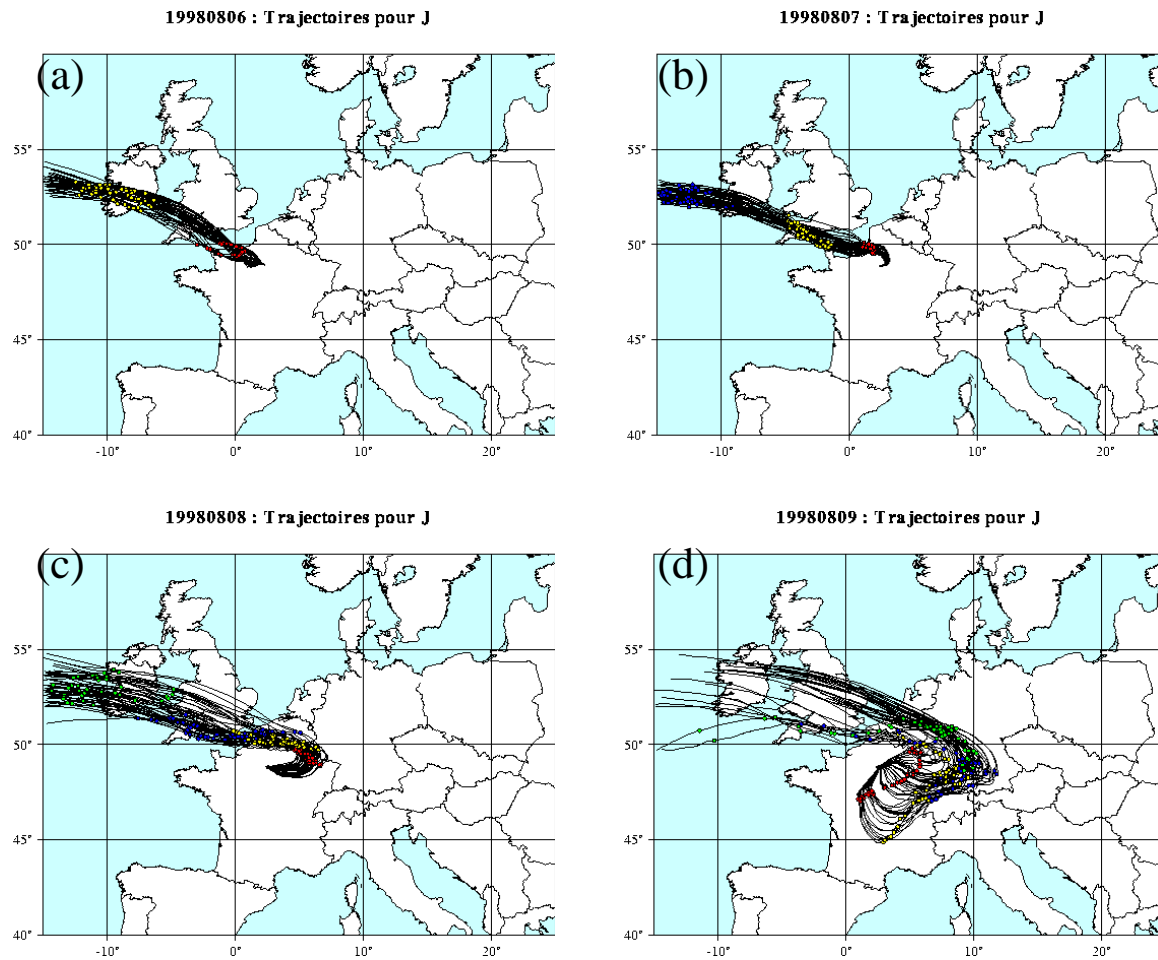


Fig. 3. Back-trajectories ending in Paris on (a) 6 August 1998, (b) 7 August 1998, (c) 8 August 1998 and (d) 9 August 1998.

Table 6. Aircraft operations during the IOP 2. 'Loops' refers to the number of circles made around Paris during the flight.

Aircraft	Day	Time period (UT)	Flight type	Loops	Levels (m)
MERLIN	7A98	0609-0711	Suburban	1	500
	7A98	1224-1540	Suburban	6	500
	8A98	0454-0832	Meso	1	900
	8A98	1232-1448	IdF	1	500
	9A98	0451-0826	Meso	1	900
	9A98	1254-1513	Suburban	5	500
AZTEC-MF	7A98	0725-0817	Urban	3	300, 900, 1500, 1650
	7A98	1444-1542	Urban	3	300, 900, 1500, 1650
	8A98	0558-0837	IdF	1	165, 330, 500
	8A98	1158-1255	Suburban	1	500
	8A98	1443-1534	Suburban	1	500
	9A98	0556-0852	IdF	1	165, 500, 1650
	9A98	1314-1625	IdF	1	165, 500, 1650
AZTEC-LSCE	7A98	0727-0919	IdF	1	300, 600, 1500
	7A98	1320-1522	IdF	1	300, 600, 1500

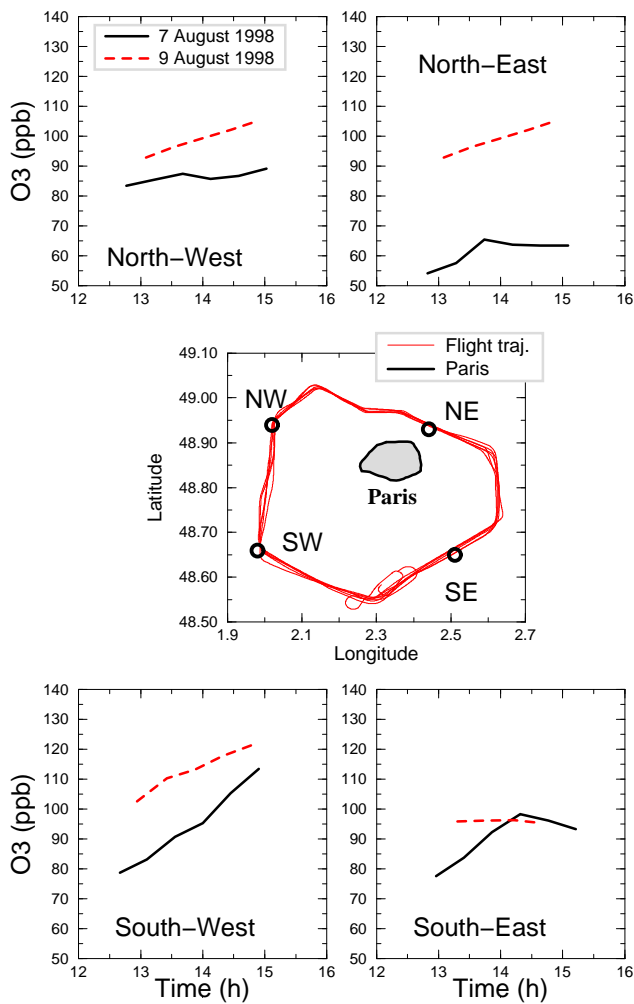


Fig. 4. Aircraft measurements of ozone, on 7A98 and 9A98, at 500 m AGL. The central box presents the city of Paris with the 'Suburban' flight trajectories superimposed. Also shown are the four representative locations selected to conduct the pollution analysis (NW, NE, SE and SW, see text for explanations). The time evolution of the ozone concentration measured in these locations are presented in the surrounding boxes.

values are of the order of 125 ppb (see Figure 8). In the urban area, concentrations are similar to upstream values.

On 8A98, the surface concentrations measured at all stations peaked around 1200 UTC which is quite unusual (as peaks are generally observed around 1500 UTC over the Paris area). A second maximum of ozone concentration is observed at the expected time (1600 UTC). In the afternoon, background values in Montgé en Goële are larger than on 7A98. The plume occurs more westward, due to light easterlies. By comparing upstream and downstream values (not shown) we conclude that the ozone production is approximately 25 ppb.

On 9A98, the evolution of the ozone concentration measured at 500 m AGL during the 5 'Agglo' flights exhibit similar trends in the NE, NW and SW locations (Figure 4), the values in the SW location being approximately 10 ppb larger. The background concentrations are much larger than on 7A98

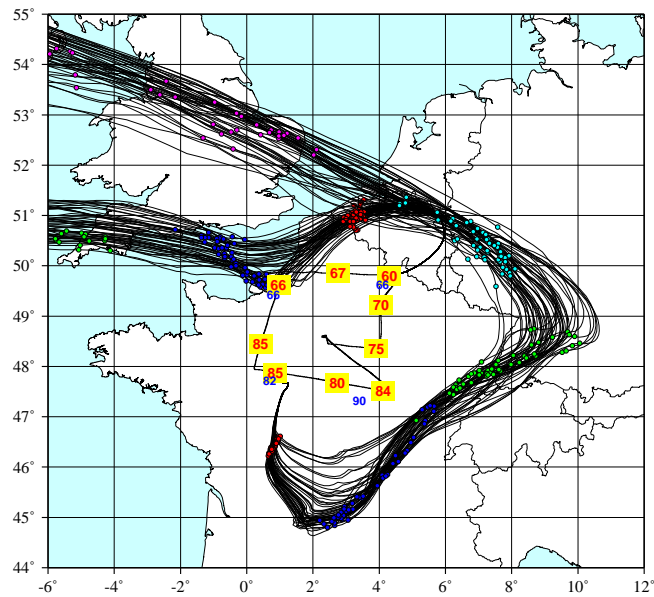


Fig. 5. Aircraft measurements performed during the Meso flight performed on 9A98 during the morning (0451-0826 UTC): boxes indicate the ozone concentrations (ppb) at 900m. The two sets of back-trajectories ending at the northeast (with $[O_3]=66\text{ppb}$) and southwest (with $[O_3]=82\text{ppb}$) corners of the Meso flight (9A98 at 0600 UTC) are estimated from back-trajectories modeling. Each point corresponds to 24h steps before the ending time, showing the air masses trajectories and the time spent above urban and/or rural areas.

and 8A98. Mean ozone concentrations differences between downwind and upwind of Paris are about 30 ppb.

Concentrations of ozone measured along the Meso flight were also very interesting (Figure 2): The flight altitude was about 900 m AGL, above the rising mixed layer, early in the morning. High concentrations (80-90 ppb) were found in the southern branch while in the northern branch concentrations were moderate (60-70 ppb). The explanation of such trends can again be given in terms of backtrajectories.

Figure 5 shows the backtrajectories, calculated with the same model as above, arriving at the northeast and southwest corners of the Meso flight trajectory. The air mass arriving at the northeast corner originates from the ocean, before spending about 24 hours above continental polluted areas (Benelux). The air mass arriving at the southwest corner has travelled above central England, Benelux, western Germany and central-east France. It has spent more than four days over continental areas, which apparently causes the higher ozone concentrations. It is noteworthy that during the two days before arrival, the air masses arriving at the southwestern corner of the Paris metropolitan area, essentially crosses rural areas, but with potentially high biogenic emissions, especially given the hot temperatures at this time (about 30-35°C). The relative importance of biogenic versus anthropogenic continental emissions will be investigated within the ESQUIF project on a modeling basis, and will be reported in a future article.

The important point here is that these measurements demonstrate that high ozone concentrations can be found over rural areas remote from important urban sources, and eventu-

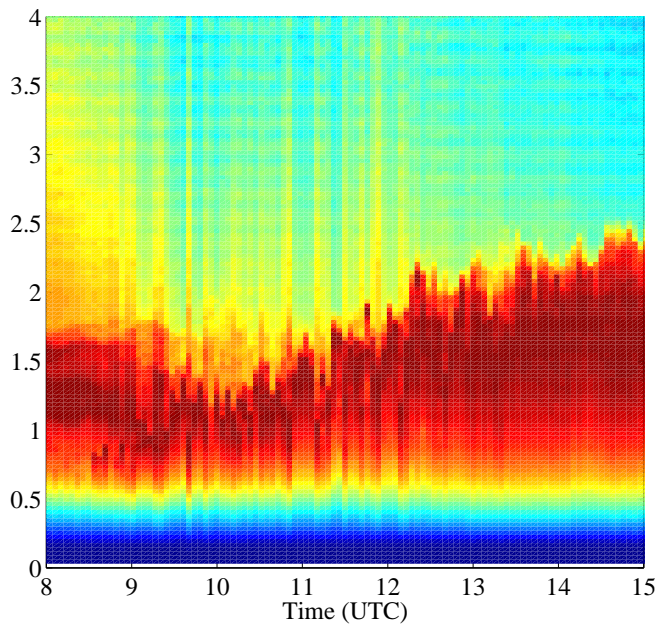


Fig. 6. Atmospheric reflectivity at a wavelength of $0.53 \mu\text{m}$ as obtained from the backscatter lidar located in Palaiseau during 8 August 1998.

ally be transported into urban areas, together with other non-oxidised precursor species.

4.2 Impact of the vertical boundary layer structure for the ozone concentrations measured over the Paris area

In addition to horizontal transport and chemical processes, the accumulation or dispersion of pollutants emitted at the surface is also driven by the convection processes within the ABL.

On 7A98, the ozone concentrations in Montgé en Goële and Paris are moderate (see previous section). However, the concentrations measured in Rambouillet at the surface and at 500 m AGL were the largest measured during the IOP. On this day, the maximum ABL depth is observed at 800 m around 1200 UTC which is much shallower than on 8A98 and 9A98 and is not representative of what is generally observed during the summer. Moreover, it remains constant for the rest of the afternoon. Meso-scale simulations were done with the Mesoscale model showing that this atypical situation was mainly due to large-scale subsidence over the Paris area. It is interesting to notice that the low ABL height was associated with the strongest ozone gradients between the city and the plume, although we cannot conclude directly that low ABL heights favours local ozone production. Indeed several effects are combined: the first IOP day, 7A98, is a Friday while the rest of the IOP occurs during the weekend, for which the emission pattern is significantly different; the second is that urban ozone concentrations can be lower when the ABL is low due to ozone titration by NO emissions; and third, in a shallow ABL higher aerosol concentrations may alter the photolysis processes in the urban area, making the city-plume gradient

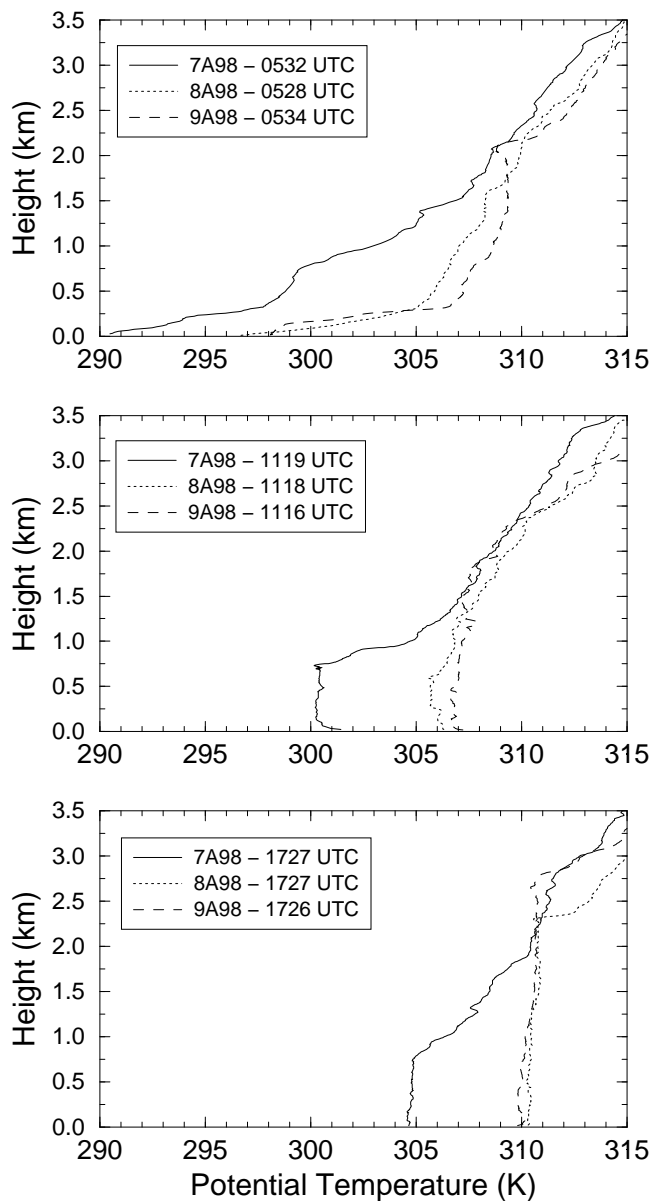


Fig. 7. Potential temperature soundings performed in Trappes, 30 km to the southwest of Paris on 7-9 August 1998.

higher. All these hypotheses will be tested using numerical modeling, and will be reported in future articles.

On 8A98, the 0830 UTC temperature sounding (not presented here) exhibited a synoptic inversion at 1300 m which was also observed on the lidar-derived atmospheric reflectivity (Figure 6) measured in Palaiseau (approximately halfway between Paris and Rambouillet). The diurnal evolution of the lidar signal shows the maintenance of an elevated polluted layer (from 600 and 1300 m) between 0800 and 0900 UTC which was decoupled from the surface by the nocturnal inversion (the height of the nocturnal temperature inversion was observed at 600 m on the 7A98 2318 UTC sounding). Around 1000 UTC, the residual layer was absorbed by the growing ABL. Surface ozone concentrations

measured in Rambouillet did not exhibit a sudden increase at this time period which could mean that the ozone concentration in the residual layer was small. The surface ozone concentration in Rambouillet increased until 1200 UTC while the ABL height rose to approximately 2300 m (from lidar data and temperature soundings) until 1700 UTC. The concentration decrease between 1200 and 1700 UTC could be explained by entrainment of low ozone background concentration into the ABL. This is in agreement with the fact that the previous day's boundary layer was rather low and that air masses entrained into the ABL thus have not encountered for the chemical ozone production the day before.

On 9A98, the ozone concentration in Rambouillet and the ABL depth increased until 1700 UTC (Figure 8 and Figure 7, respectively). However, the previous day's ABL was also high, explaining high ozone concentrations found at high altitude. Ozone peaks were much later than the previous day, since the dilution effect was not as effective on this day. We also note an increase, during the afternoon, in the background ozone (see on Figure 8 the Montgé-en-Goële station), which could also explain the general increase of all ozone concentrations at the end of the afternoon. Again, definitive conclusions can only be given after numerical simulation of the episode using both regional and continental scale models.

5 IOP2 of ESQUIF: Model analysis

The first goal of analysis modeling is to determine if CTM models are able to reproduce a complex pollution event as the one observed during IOP2. The simulation was performed from 7A98 (0000 UTC) to 9A98 (2300 UTC). In order to account for small-scale surface heterogeneities, ECMWF profiles were interpolated to the surface using observed temperature and pressure data (from surface stations). Ozone concentrations resulting from the three-dimensional simulations are compared to those measured by surface stations on an hourly basis. In Figure 8, comparisons are displayed for Paris (top), Montgé-en-Goële, northeast of Paris (middle), and Rambouillet, southwest of Paris (bottom). For the three locations, the agreement is fair over the three days. In Paris, on 7A98 and 8A98, concentrations are representative of the averaged observed concentrations, and on 9A98, in Paris, one notes a model underestimation at the end of the day. In Montgé-en-Goële, the model fits the observations well which are representative of background ozone advected over the whole simulated domain. Over Rambouillet, on 8A98 and 9A98, the model fits well the surface ozone concentrations, but some discrepancies appear on 7A98 where the plume intensity is underestimated. In order to understand this underestimation, we turn to Figure 9 which displays a map of simulated ozone (in ppb, at $z=500$ m AGL, on 7A98 and at 1500 UTC) over the Paris area.

These simulated ozone values are compared to aircraft ozone measurements, performed at the same period. The plume amplitude is well reproduced, but it is slightly shifted southward compared to observations. The discrepancies between modelled and observed values may be explained in terms of the mean meteorological forcing fields used: the ECMWF inputs correspond to the mean flow over the whole domain, without topography at small scale. Errors induced

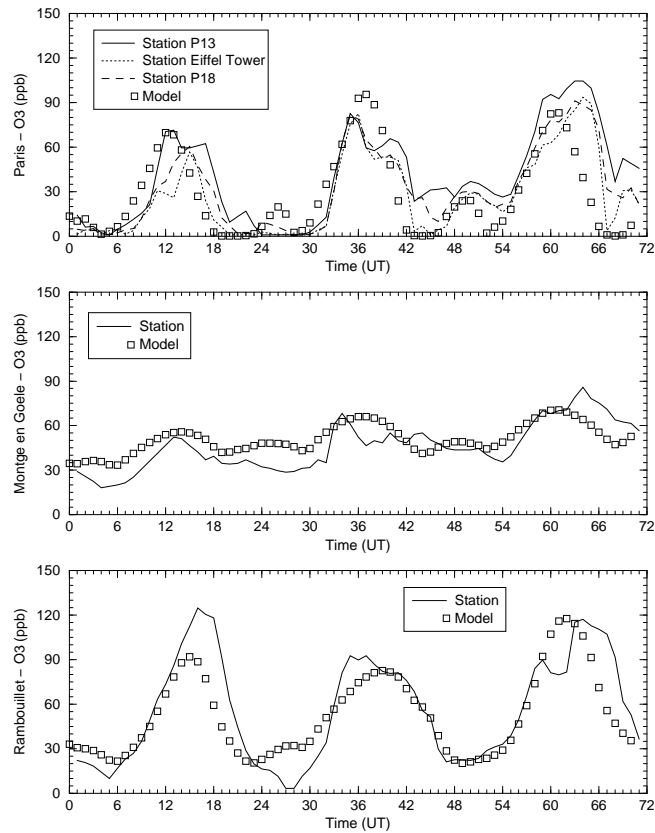


Fig. 8. Surface ozone concentrations (ppb) measurements by AIR-PARIF from 7 to 9 August 1998 (from 0 to 72h) in Paris (top), Montgé-en-Goële (middle), and Rambouillet (bottom). The Montgé-en-Goële and Rambouillet suburban stations are located to the northeast and southwest of Paris, respectively. Results from the CTM model are added at each location for comparison with in-situ measurements

using these meteorological fields may be of the order of a few percent for wind speed and direction. The next step to improve the analyses performed with POLLUTO will be to use simulations made by a fully mesoscale model, such as Meso-NH in order to account for small-scale topography and more accurate turbulent effects. This step is currently in preparation.

6 Summary and conclusion

The objectives and the experimental set-up of the ESQUIF project have been presented. Numerous ground and airborne measurements have been acquired in the course of 10 IOPs, which cover a variety of meteorological situations leading to air pollution in the region of Paris. This project is part of a series of European projects dedicated to atmospheric pollution which have been undertaken recently. With respect to the other projects, one outstanding goal of the ESQUIF project is to implement a thorough data base which would include the measurements as well as model simulations. This data base exists (<http://www.lmd.ens.fr/ESQUIF/>) and will be available

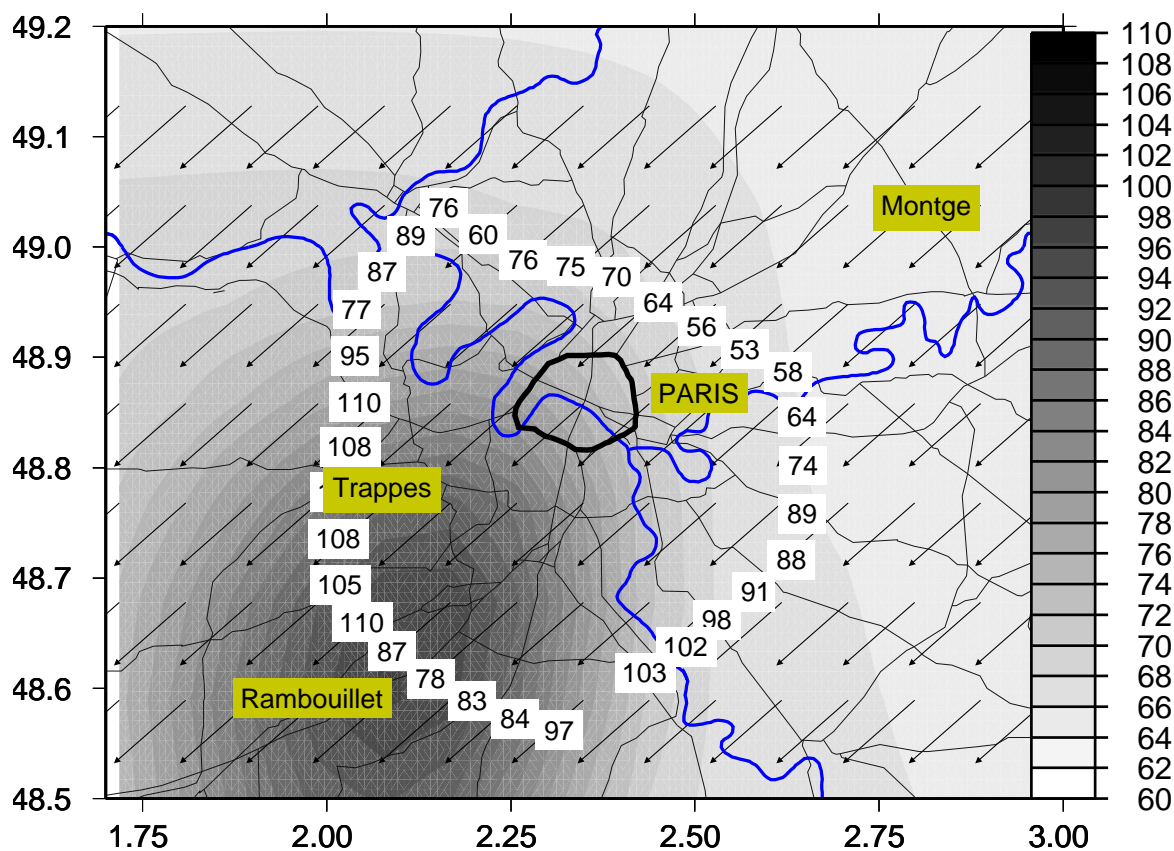


Fig. 9. Measurements and model comparison for $[O_3]$ (ppb) concentrations on 7A98 at 500 m AGL and 1500 UTC. The city of Paris is at the center of the model domain, and the concentrations are representative of the vertical box model at $z=500\text{m}$. The aircraft measurements were performed during a 'Suburban' flight (7A98, from 1430 to 1500 UTC, with the MERLIN aircraft, see Table 6) and $[O_3]$ (ppb) measurements are superimposed along the flight trajectory. Wind vectors are issued from ECMWF data.

shortly to the scientific community interested on atmospheric pollution studies.

In this paper, we have presented preliminary results obtained during the second IOP of ESQUIF, in August 1998. The study conducted here demonstrates that only a continental-scale approach enables a thorough understanding of atmospheric photochemical pollution over a strongly urbanized area such as Paris. Indeed, strong pollution events were diagnosed to be influenced by dynamical and chemical processes at different scales. A major reason for this is certainly the absence of marked orography. For the situations documented here, it was shown that, besides surface emissions, the major factors driving pollutants concentrations were continental-scale transport and convective activity.

The main result of this study is to show that, combining modeling and experimentation, we could identify the contribution of local production versus regional transport to the ozone peak. Using this combined approach, we have shown that on 7A98, the pollution event is mainly due to local production, whereas on 9A98, high ozone concentrations are due both to local production and regional ozone transport.

Acknowledgments

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