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Flora Toussaint, Patrick Chazette, Hariliva Randriamiarisoa, Joseph Sanak, Pierre Couvert, Bernard Bonsang, Patricio Moral, Sébastien Berthier, Daniel Martin, Franck Toussaint

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**N . A . I**

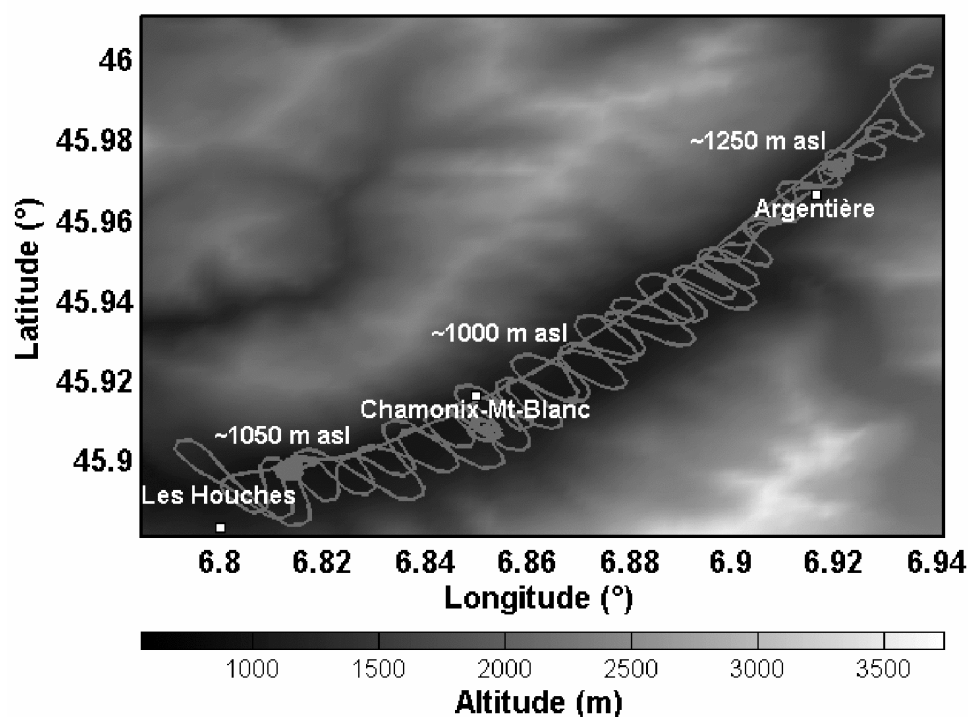
NOTES DES ACTIVITÉS INSTRUMENTALES  
INSTRUMENTS - EXPÉRIENCES - OBSERVATIONS

THREE DIMENSIONAL SURVEY OF POLLUTION DURING WINTER  
IN THE FRENCH ALPS CHAMONIX VALLEY FROM AN ULA

Patrick Chazette<sup>1</sup>, Hariliva Randriamiarisoa<sup>1</sup>, Joseph Sanak<sup>1</sup>, Pierre Couvert<sup>1</sup>, Bernard  
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# NOTES DES ACTIVITÉS INSTRUMENTALES DE L'IPSL

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# Three dimensional survey of pollution during winter in the French Alps Chamonix Valley from an ULA

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## Abstract

We developed and installed a scientific payload onboard an ultra-light aircraft (ULA) dedicated to ozone, nitrogen oxides, and aerosol scattering profilers to study the pollution events in the Chamonix Valley (French Alps). Meteorological parameters (temperature, pressure, relative humidity) and altimetry (GPS measurements) were recorded simultaneously. The experiment undertaken in January 2003 during daytime allowed us to document, for the first time, the pollution events on horizontal and vertical scales during typical winter situations in the Chamonix Valley. Our results show a strong inversion of temperature that traps the pollutants close to the surface. This shallow inversion layer corresponding to the planetary boundary layer seems to remain within a range of about  $150\pm 50$  m above the ground level and is closely associated to a minimum of ozone mixing ratio linked to an increase of nitrogen oxides (NO and NO<sub>x</sub>). The concentrations of pollutants are thus shown to be closely linked to the proximity of their sources. A transition layer with the free troposphere may also be observed above the PBL.

Keywords: pollution, airborne measurements, Alpines valleys, POVA, ultra-light aircraft.

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## 1. Introduction

Air quality management in both urban and suburban areas and its spreading to a regional scale is becoming a major preoccupation of our modern society (i.e. Lelieveld et al., 2001). Pollution impacts on both ecological equilibrium and public health. Various studies over the last twenty years have led to a better knowledge of the physico-chemical processes leading to pollution events in urban and rural areas particularly where ozone (i.e. Menut et al., 2000, Vautard et al., 2001) and particles (i.e. Odum et al., 1997; Forstner et al., 1997) formations are concerned. Nevertheless, the lack of observations to constrain models remains a major limitation. Continuous in situ observations leading to extensive data sets and better resolved time and space measurements are needed for the understanding of the physics and the chemistry involved in regional pollution processes.

Traffic is one of the main sources of photo-oxidant pollution and aerosols (i.e. Vautard et al., 2003). The nonlinear character of chemical transformations occurring in pollution plumes may affect and make difficult the setting up of regulation policies concerning the different components of pollution emissions. The tri-dimensional distribution of pollutants is one of the important informations that must be taken into account to assess their environmental impacts. Numerous campaigns have therefore been conducted in order to investigate the 3-D distributions of pollution plumes as well as their temporal evolution. On a regional scale, major cities have been extensively investigated : Paris (France) (Menut et al., 2000; Vautard et al., 2003), Los-Angeles (California) (i.e. Lurmann et al., 1997), Athenes (Greece) (i.e. Eleftheriadis et al., 1998) and Rio (Brazil) (Grosjean et al., 1990). Larger scale experiments, involving a wider palette of pollution sources, have been conducted over continents and oceans such as the Aerosol Characterization Experiment (ACE-2)

(Flamant et al., 2000) or the INDIan Ocean EXperiment (INDOEX) ( Lelieveld et al., 2001; Chazette, 2003).

The main goal of the “POllution dans les Vallées Alpines” (POVA) program is to study the pollution in the Alps valleys, to identify the sources and characterize the dispersion of pollutants. This program was initiated because of the ecological and public health problems raised by the reopening of the Mont-Blanc tunnel to truck traffic. The Mont-Blanc tunnel had been closed after a serious accident which occurred in March 1999. The program POVA seeks to evaluate the respective contributions of the various types of emissions (biogenic and anthropogenic) in the chemical processes involving gas and particulate atmospheric species, and to determine their variability according to the environmental conditions. Alpine valleys are a very peculiar framework for atmosphere chemistry studies, due to the spatial concentration of the precursor emissions in steep-sided valleys and to the specific dynamical and physico-chemical conditions (convective activity, valley breezes and orographical forcing, frequency of temperature inversions, intense solar radiation related to altitude) which vary from one season to another.

The concept of a relatively simple payload to monitor the vertical distribution of atmospheric aerosols based on in situ scattering measurements has been validated by Rosen and Kjome (1991), Rosen et al., (1997) and reused by Dulac et al. (2002). Measurements performed from ultra-light aircraft (ULA) have been initiated by Junkerman (2001) during the ESCOMPTE campaign in June-July 2000. They have proved the scientific interest of such an approach for sampling pollution plumes in a complex topography. To better understand the role of both gas and aerosols in mountain valleys pollution events, it is necessary to follow their evolutions over time and space in the atmospheric column. Using a ULA to perform airborne

measurements seemed thus particularly appropriate to investigate the steep-sided alpine valleys at different altitude levels. The first experimental campaign using our ULA took place from January 14<sup>th</sup> to 22<sup>nd</sup> 2003 in the Chamonix Valley.

The first results obtained during this winter campaign are presented. The instruments used onboard the ULA are described together with the different flight plans. Both vertical and horizontal structures observed in winter conditions are then presented and the influences on the pollutants are discussed.

## **2. Experimental setup**

The ULA used during our experiment was built by Air Creation company ([www.aircreation.fr](http://www.aircreation.fr)). The maximum payload is approximately 230 kg (including the pilot) and we boarded close to 80 kg of scientific payload. The cruise speed ranges from 75 to 130 km/h with a maximal autonomy of 3 hours. The ULA can explore an altitude range between 0 and ~4.5 km above sea level (asl). The advantages of such an ULA are its maneuverability in atmospheric volumes of low dimension and its low speed of flight which minimizes the isokinetic problems involved in aerosols measurements.

### **2.1 Payload**

The specific payload onboard the ULA is summarized in Table 1. Ozone, nitrogen oxides (NO and NO<sub>x</sub>) and carbon monoxide concentrations were recorded with the new compact instrument SYIMCA. Simultaneously, the scattering coefficient of aerosols, temperature, relative humidity, atmospheric pressure and position (by GPS) were measured.

Table 1: Payload of the ULA with its specific characteristics

Instrument	Accuracy	Geophysical parameter
SYIMCA O <sub>3</sub>	2 ppbv	O <sub>3</sub> mixing ratio
SYIMCA NO	200 pptv	NO mixing ratio
SYIMCA NO <sub>2</sub>	500 pptv	NO <sub>2</sub> mixing ratio
SYIMCA CO	1 ppmv	CO mixing ratio
PdRam	<10%	□ <sub>a</sub>
Meteorological parameters	~0.5 K 1% ~1 hPa	Temperature Relative humidity Pressure

### 2.1.1 The SYIMCA analyser

This multigaz analyser, designed by « Environnement S.A. » as a mobile equipment, simultaneously measure O<sub>3</sub>, NO, NO<sub>2</sub> and CO concentrations on a continuous real time basis. It is enclosed in a 60 x 40 x 20 cm box weighting 23 kg. An external pump ensures stabilized 3.2 l/mn flow rate through the analysers. Automatic corrections for temperature, pressure and flow rate are made by software. Data are collected by an external microcomputer on a 5 second basis through a RS232 connection together with the other on board measurements. Electrical power is supplied by a small generator (Honda, 900 VA).

Calibrations were performed before and controlled after the campaign with a “Thermo-Environmental” 49PS model for ozone, and with calibrated gases for the nitrogen oxides and carbon-monoxide. The accuracies of measurements are given in Table 1 for each gas component. The detection limits are equal to the given measurement accuracies thus permitting the determination of the pollution level without ambiguity.

### 2.1.2 The Personal DataRam (PdRam)

Anthropogenic aerosol scattering is a passive tracer of both the inversion layer and the planetary boundary layer (i.e. Flamant et al., 2000; Chazette, 2003) but it is highly dependant on humidity due to water absorption/condensation. Based on these



concepts, we interfaced a Miniature Real-time Aerosol Monitor (PdRam), manufactured by M.I.E. Inc. (Billerica, Massachusetts), with a meteorological pressure-temperature-humidity (PTU) monitor. The PdRam is a small portable nephelometer-type instrument working at 880 nm. Previously used onboard a balloon, it is well documented in Dulac et al. (2002). For our application, PdRam is calibrated at ground level using the 550 nm wavelength of a laboratory calibrated TSI-nephelometer (Bodhaine et al., 1991; Veeffkind et al., 1996). Since the light scattered by the particles within the measurement chamber is only integrated between  $7^\circ$  and  $170^\circ$ , the results provided by the nephelometer underestimate the real scattering coefficients by  $\sim 4\text{-}5\%$  for anthropogenic aerosols after molecular scattering correction. After calibration, the mass concentration  $M_a$  provided by the DataRam leads to a scattering coefficient  $\sigma$  (in  $\text{km}^{-1}$ ) given by  $0.0023 M_a + 0.0018$  for relative humidity RH lower than 70%. The residual accuracy is about  $0.01 \text{ km}^{-1}$ . If the relative humidity is greater than 70%, a correction factor must be applied following  $(880/550)^{a + a_{RH}}$  where  $a$  and  $a_{RH}$  are the spectral dependency of the scattering coefficient respectively for  $\text{RH} < 70\%$  and  $\text{RH} > 70\%$ . For instance, the correction factor has been assessed at the ground level to be  $\sim 1.1$  for  $\text{RH} = 80\%$ .

## 2.2 Flight plans

The flight plans were defined to explore the valley in 3 dimensions at different hours during daytime. For each ULA flight, measurements were performed at different horizontal levels, typically within layers 150-200, 250-300 and 350-400 m above the valley bottom (avb). We have explored the lower and upper parts of the valley to follow the pollutant spreads and examples of flights plans are given in Figure 1. During the flights, the ULA stayed far enough from the mountains slopes to limit their influence. Measurements are thus more representative of the valley core.

Each horizontal level was explored through zigzag flight leading to a good mapping of the layer atmospheric characteristics. Specific vertical profiles were acquired over characteristic areas by means of helix trajectories. The atmospheric column was thus explored at different locations close to the cities highlighted in Figure 1.

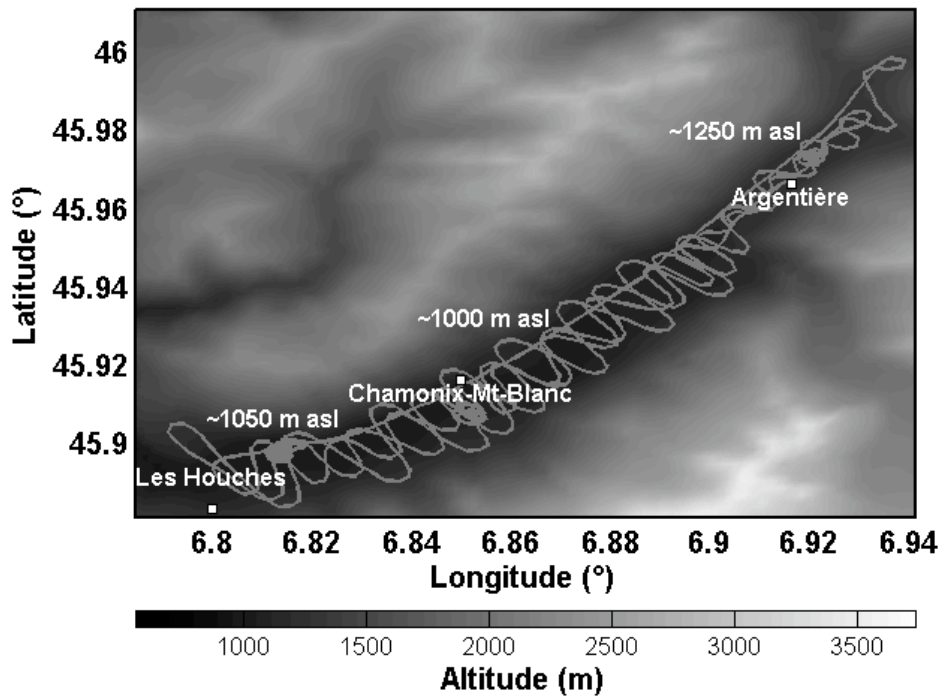


Figure 1: Flight plan type performed by the ULA during the experiment in the Chamonix Valley on January 17<sup>th</sup> 2003.

Flights were possible only during daytime and in cloud free condition. The measurement periods are listed in Table 2. Both morning and afternoon measurements were performed to follow the temporal evolution of the pollutant concentrations with respect to vertical stability of the low troposphere. The concentrations of CO were not significant enough due to a calibration problem and have not been used hereafter. The PdRam scatterometer was not operational January 19<sup>th</sup> and 20<sup>th</sup>, due to electronic failure.

Table 2: Timetable of measurements from ULA for the Chamonix Valley.

Flight number	Date	Takeoff (UT)	Landing (UT)	Flight number	Date	Takeoff (UT)	Landing (UT)
1	15	9h15	10h15	12	17	16h45	17h10
2	15	10h30	12h10	13	18	8h40	10h40
3	15	14h15	16h00	14	18	10h50	11h50
4	16	8h45	9h45	15	18	14h00	16h00
5	16	10h00	11h10	16	19	9h00	10h05
6	16	14h20	15h20	17	19	10h15	11h45
7	16	15h30	16h45	18	19	14h30	16h00
8	17	10h15	11h05	19	20	9h00	10h45
9	17	11h15	12h00	20	20	13h45	14h00
10	17	14h20	15h05	21	20	14h10	14h15
11	17	15h10	16h30	22	20	14h20	14h45

### 3. Discussion

#### 3.1 Meteorological situation

##### a. Synoptic conditions

The majority of the observations were done under strong anticyclonic conditions with a surface pressure at sea level of about 1030 hPa. This blocking system is very favorable for occurrence of marked inversion layer close to the surface which may trap the pollutants within less than one hundred meters above ground in winter period. If the synoptic wind remains sufficiently weak, it will not disturb the air circulation in the bottom of the valley. After January 20<sup>th</sup>, a low pressure system centered on Iceland and generating a warm front brought foehn winds which disturbed the air circulation in the valley. Rain followed by snow ended the atmospheric measurements.

## b. Local conditions

Local circulation in mountain valleys is mainly influenced by topography and the Planetary Boundary Layer (PBL) processes display peculiar features associated with various constraints due to horography. Synoptic modeling is thus inevitably limited and must be supported by assumptions concerning ascents, subsidences, as well as history of the air masses. During the experiment, dynamic air flow measurements were not possible due to experimental payload complexity and cost. Nevertheless, the typical winter conditions experienced during the campaign, with a mean horizontal surface wind speed always lower than  $2 \text{ ms}^{-1}$  and a strong temperature inversion due to extended snow cover, support the hypothesis of a strong concentration of the pollutants in the valley bottom.

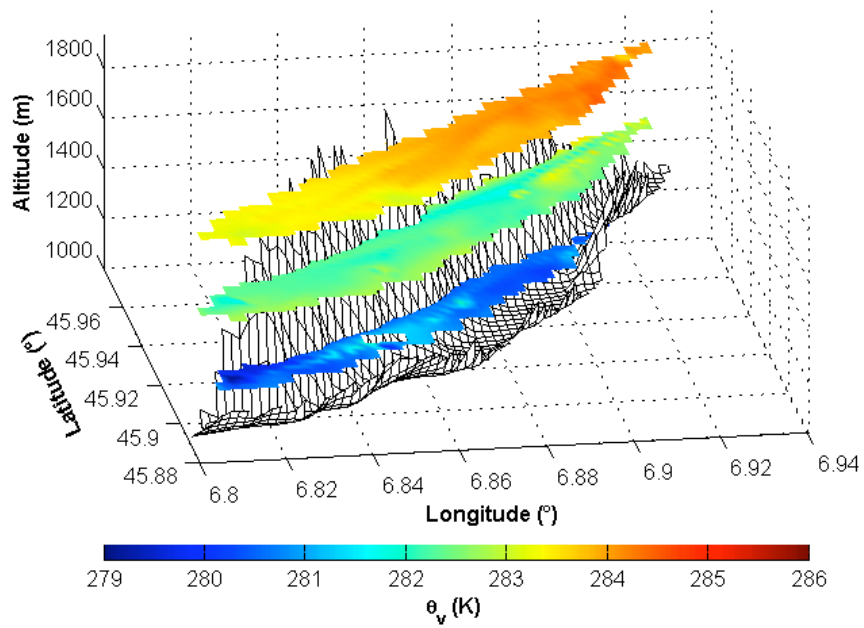
Even during winter, solar irradiance could play a role because of the heat gradient which can exist between the enlightened and the shaded valley faces. Specific convective processes to each slope can thus exist according to their orientation with respect to the sun. Morning anabatic and evening catabatic breezes may then participate in the mixing of pollutants such as ozone, produced on a regional scale, with the air of the valley bottom. However, for the Chamonix Valley, oriented along a NNE-SSW axis, such processes should be attenuated with the exception of its northern extremity, around Argentière, where slopes are southerly oriented.

During the day to day ULA flights, the meteorological conditions were fairly stable and we have measured rather similar vertical and horizontal structures and concentrations. We have mainly selected January 18 where both daytime and spatial ranges were well sampled and truck traffic was effective on the tunnel highway to illustrate the three-dimensional spread of pollutants.

### 3.2 The vertical atmospheric structure

Figure 2 shows the recomposed plans of virtual potential temperature  $\theta_v$  from ULA flights over the valley between Les Houches and Argentière (Figure 1). For each plan, the flight altitude is constant relative to sea level as mentioned above. A strong vertical gradient of  $\theta_v$  is observed. As expected, it is representative of a well established vertical stability close to the valley bottom. A very small horizontal variability of temperature can be noted for each flight level. Nevertheless, the temporal evolution between the morning and after midday is significant. It is related to the radiative heating of the valley bottom air with a small reduction in vertical stability due to the vertical convection development during the afternoon. Small increases of  $\theta_v$  can be detected around  $6.85^\circ$ , before  $6.82^\circ$  and after  $6.87^\circ$  of longitude, corresponding to the location of Chamonix, Les Houches and Argentière where an urban heat island effect may occur. The RH behavior (not shown) is in agreement with that of  $\theta_v$ ; a rapid decrease is observed between mid-morning and early afternoon (from 60% to 45% at ground level) associated with the temperature enhancement. As expected, the highest values of the aerosol scattering coefficient  $\sigma$ , given in Figure 3, are observed at surface level. The recorded level remains very weak ( $< 0.03 \text{ km}^{-1}$ ) and sharply decreases with altitude. Higher values are observed over urbanized zones like Les Houches and near Argentière (values over Chamonix are missing).

a)



b)

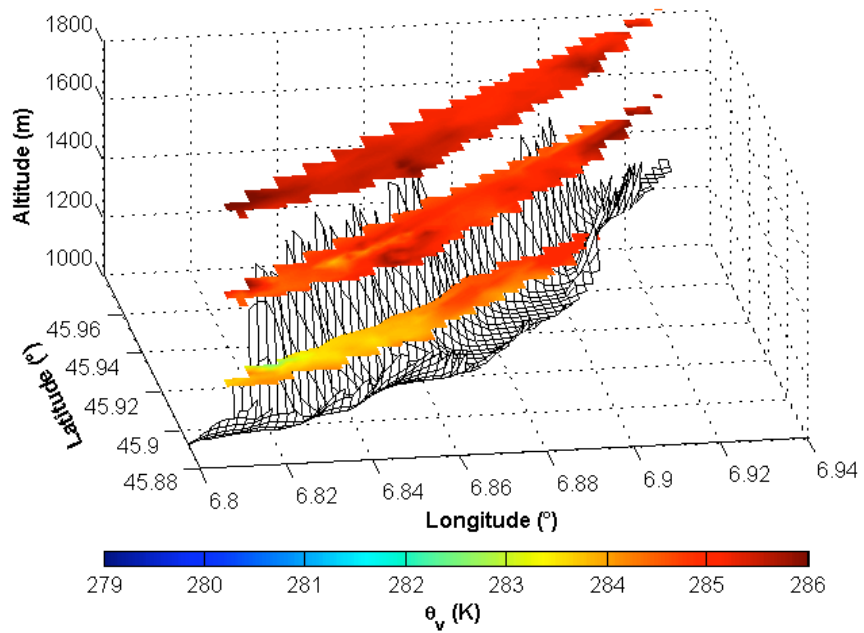
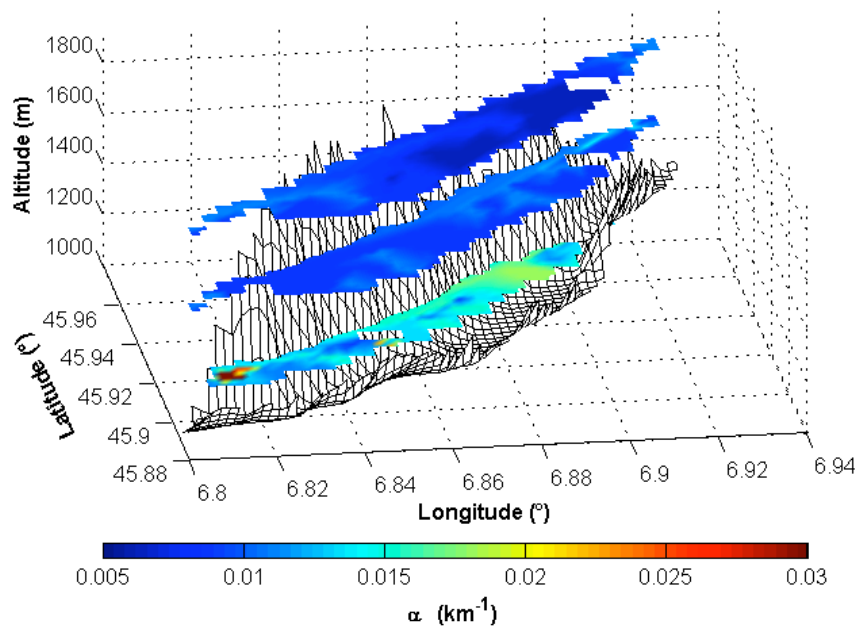


Figure 2: Virtual potential temperature  $\theta_v$  from ULA flights over the valley, between Les Houches and Argentière, for January 18<sup>th</sup>. Figure a is for 8h40-10h40 UT and figure b for 14h00-16h00 UT. The 3 flight levels are at ~1150, ~1300 and ~1450 m asl.

a)



b)

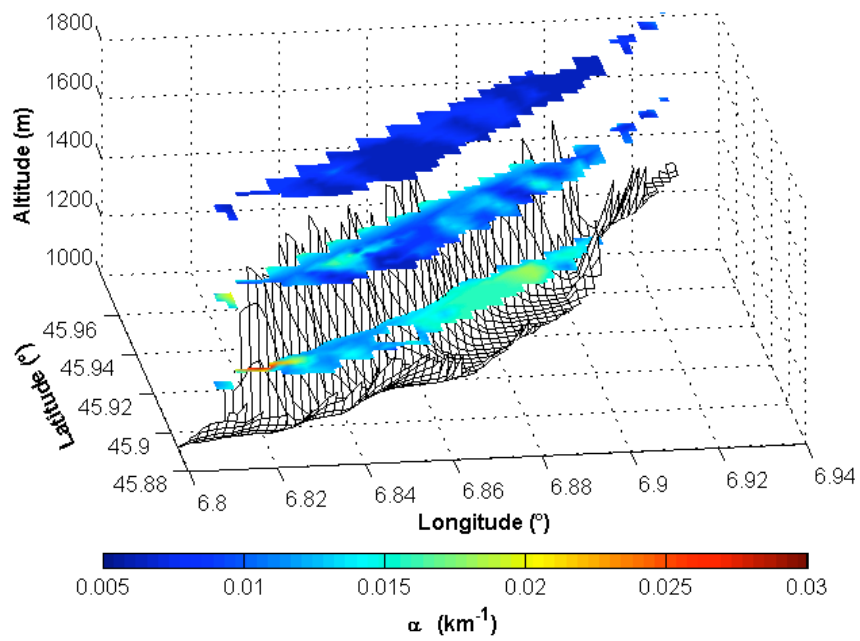


Figure 3: Same as Figure 2 for aerosol scattering coefficient  $\alpha$ .

The structures described above are representative of a persistent inversion layer throughout the day. It is linked to the low surface temperature (between  $-10^{\circ}$  and  $3^{\circ}\text{C}$ ) and a snow cover over the whole valley. The pollutants are thus trapped near their sources within the PBL layer, between 0 and 150 m avb. This situation persisted throughout the measurement period as illustrated in the Figure 4 where the mean vertical profiles of  $\sigma_v$  are given for each measurement day. For both morning and afternoon flights, the whole valley data are averaged within 50 m altitude bins. Above the PBL, a transition layer could be observed during the afternoon, between  $\sim 1100$  and  $\sim 1400 \pm 100$  m asl (Figure 5b - January 16<sup>th</sup> and 17<sup>th</sup> are the most probing examples). The top of this layer was not accurately determined due to flight altitude limitations for pilot safety in winter conditions. Such a layer may be linked to the “mountain injection layer” identified by Graber et al. (2000) in studies performed on the transport of pollutants in the Alps valleys during summer. This transition layer is more difficult to identify on the virtual potential temperature profiles which sometime highlight a multi-layer structure due to the high vertical stability, as for January 19<sup>th</sup>. The aerosol scattering coefficient is significantly higher in the bottom part of the profiles for morning measurements. This may be due to weaker mixing efficiency following the altitude. The exception of January 16<sup>th</sup> is associated with lower temperatures throughout the day and weaker atmospheric stability. Hence, in the morning, the particles could be mixed with higher altitude air masses and thus be less concentrated at the ground level (Figure 5a). During the afternoon,  $\sigma$  increases significantly from  $\sim 0.05$  to  $\sim 0.12 \text{ km}^{-1}$  at the ground level. This increase is not due to a significant modification of the relative humidity which remains very stable for this day with values between 70 and 75% for the 1000-1500 m asl range. Influences of synoptic or regional air masses have been discarded by the analysis of backtrajectories



performed on 3 days starting at 18h GMT (courtesy of NOAA Air Resources Laboratory <http://www.arl.noaa.gov>) and so the aerosol origin must be local.

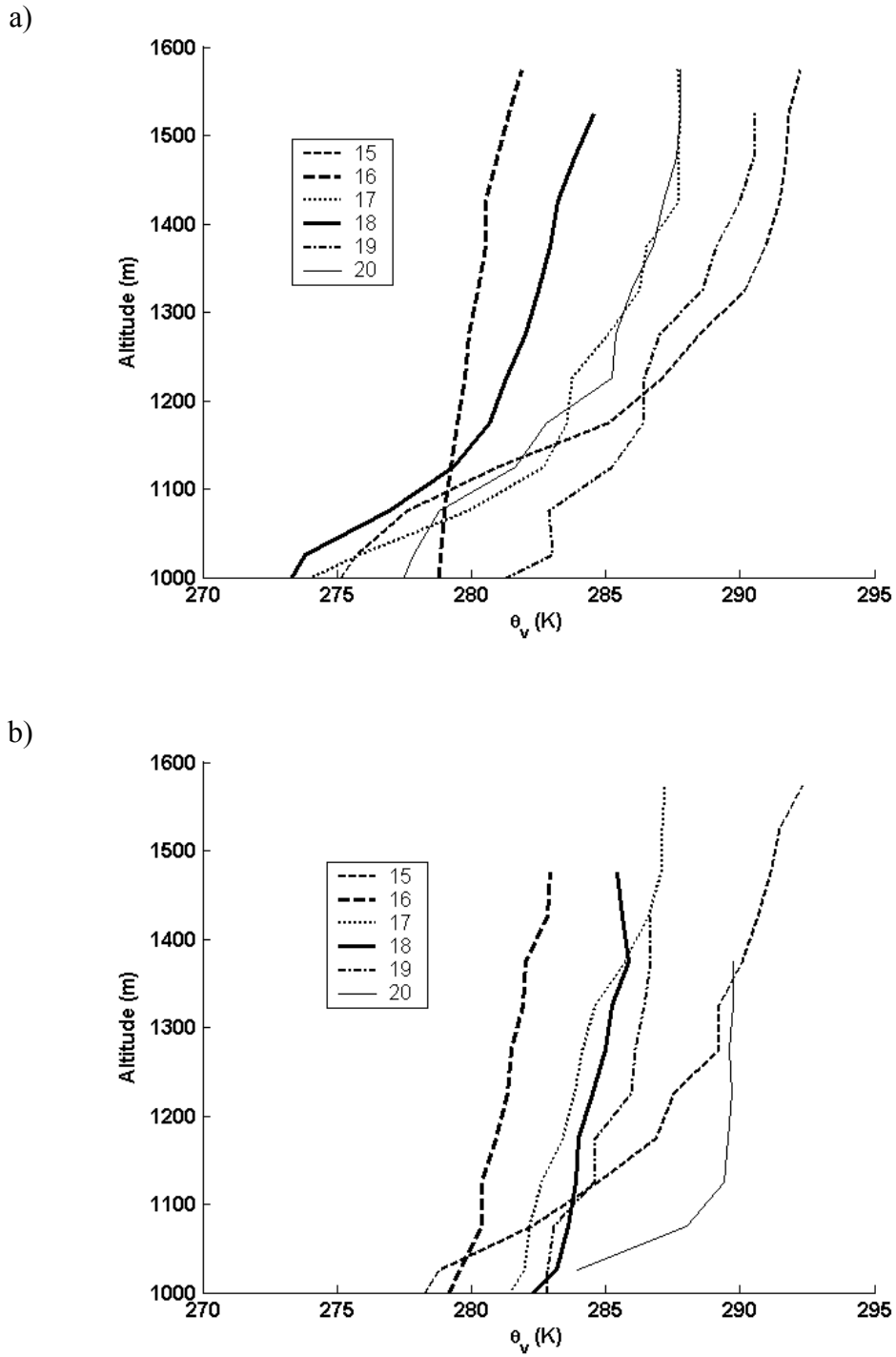
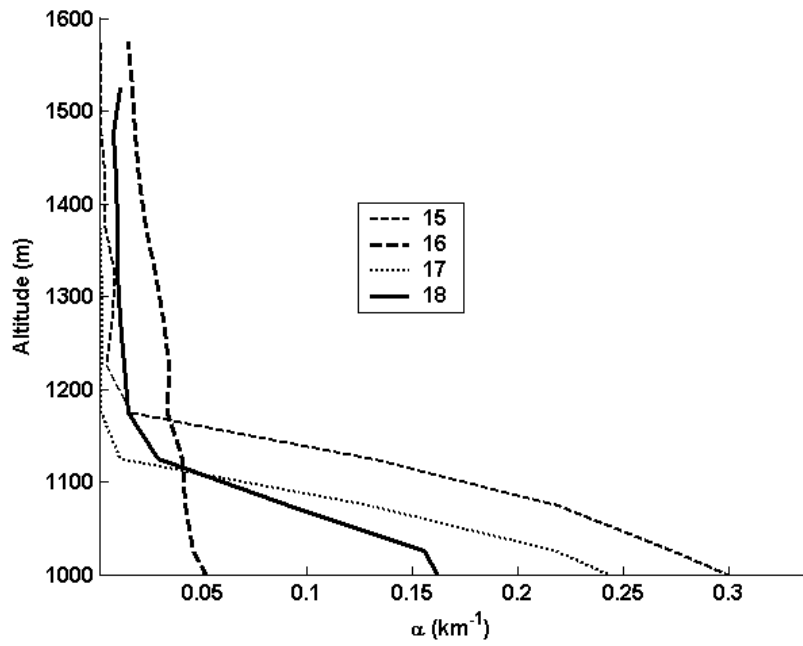


Figure 4: Day to day mean vertical profiles of virtual potential temperature  $\theta_v$  for the morning (a) and the afternoon (b).

a)



b)

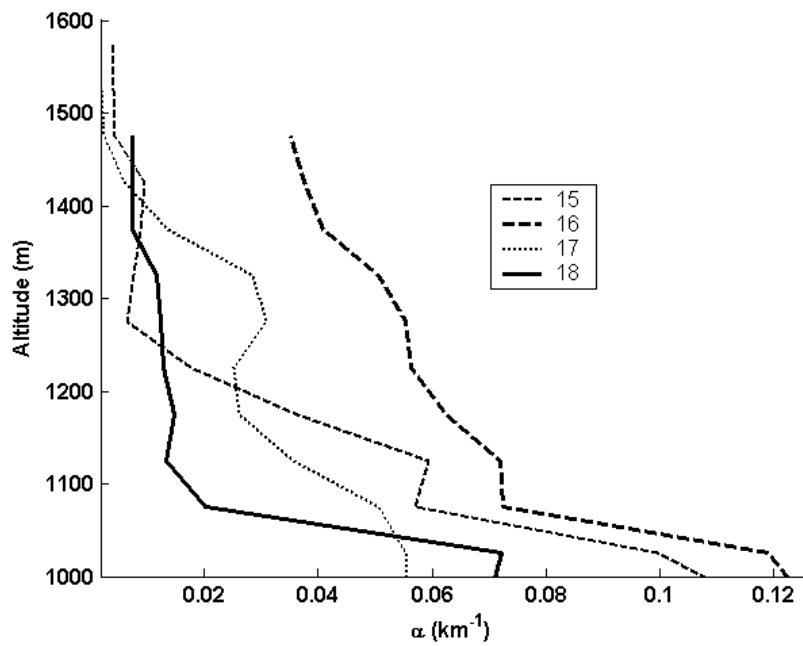
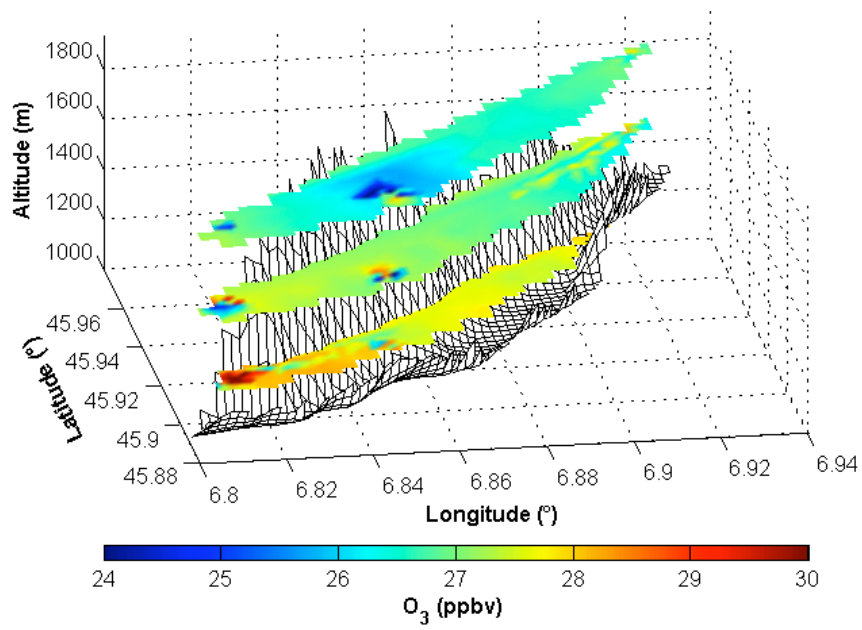


Figure 5: Same as Figure 4 for aerosol scattering coefficient  $\alpha$  (note the scale change between morning and afternoon).

### 3.3 Impact on pollutants

Measurements of NO, NO<sub>x</sub> and O<sub>3</sub> confirm the preceding conclusions relative to the vertical structure. The pollutants generally present more important concentrations in the lower level, close to the sources. As shown in Figure 6, the overall ozone concentration is lower during the morning and increases during the afternoon because of the emissions of ozone precursors (nitrogen-oxide, volatile organic compound) mainly emitted by automobile traffic and urban heating. Above Chamonix (~6.85° of longitude), one clearly observe the urban heat island effect, from the ground up to 400 m avb. The morning ozone concentration is minimal in the upper level, right above Chamonix, because of its titration, during the night, by ascending nitrogen oxides (Figure 7). It increases during the day, for the 3 levels, following the human activities. Similar observations are made above Les Houches (longitude lower than 6.82°). Strong concentrations of NO (not shown) are also measured in the lower level near Les Houches with values higher than 10 ppbv. The values of NO<sub>x</sub> (NO + NO<sub>2</sub>) are also very important close to the ground throughout the day as shown in Figure 7. They largely exceed 10 ppbv, in the afternoon, in the southern part of the valley where the tunnel highway is located. .

a)



b)

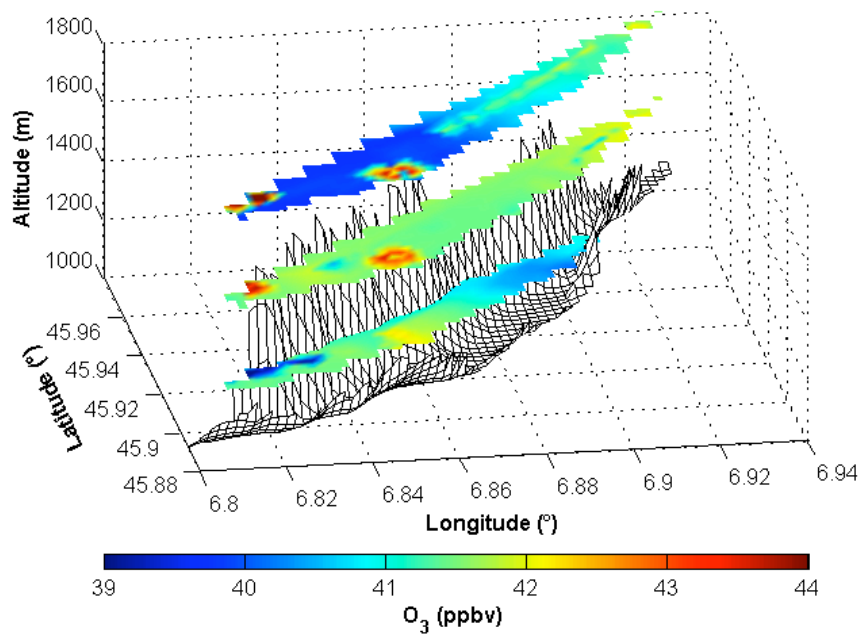
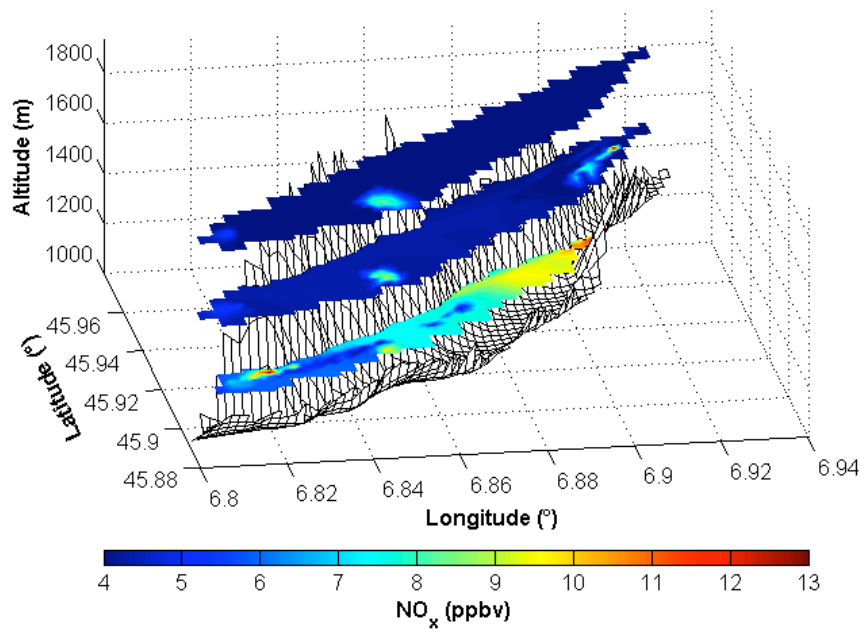


Figure 6: Same as Figure 2 for O<sub>3</sub> (note the scale change between morning and afternoon).

a)



b)

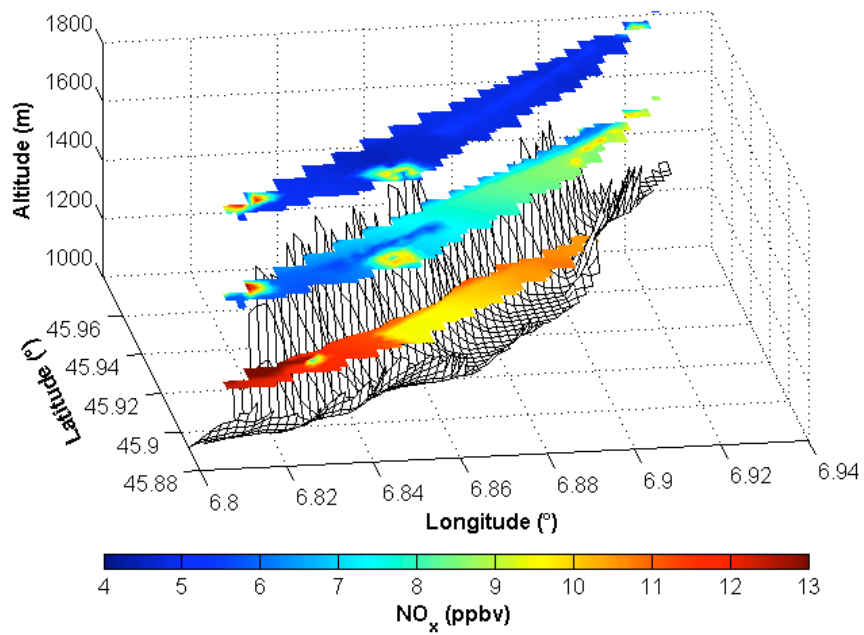


Figure 7: Same as Figure 2 for NO<sub>x</sub>.

The day to day evolution of the ozone concentrations is given in Figure 8. Lower ozone concentration can be observed during the morning for January 15, 16 and 18 above PBL. For these days the synoptic subsidence is very weak with air masses from West-Northwest between 800 and 5000 m asl, and small quantity of the free troposphere ozone may penetrate into the valley. Nevertheless, the synoptic transport cannot alone explain the behavior shown here. For instance, the nitrogen oxides concentrations are very important for January 16<sup>th</sup> compared to the other days (Figure 9), with low NO concentration (<2 ppbv) above the PBL for all days (not shown). A hypothesis is that the low temperature present during this morning may inhibit the ozone chemistry keeping nitrogen oxide in an oxidized form. During the afternoon, the ozone vertical profile of January 16<sup>th</sup> is weak (<20 ppbv) with high values of NO<sub>x</sub> but similar low values of NO as for the other days. The temperature remained very low throughout the day (Figure 4) and the ozone chemistry was probably limited. A similar analysis could be made for January 15 where the very high stability of the low troposphere traps pollutants close to the ground. NO concentrations then stay high (> 40 ppbv) leading to a low ozone concentrations during the afternoon.

The PBL top location is confirmed to be under 150 m with a transition layer just above. This latter layer is more evident on the NO<sub>x</sub> profiles, mainly for January 16 and 17. It has been qualified as an injection layer by Graber et al (2000) because it receives the air masses advected from the PBL. They situated this layer between 1500 and 4000 m asl during summertime and it is not surprising that its altitude decreases during wintertime.

Finally, it is interesting to note that the ozone profile of January 19<sup>th</sup> is not specific and corresponds to a Sunday without truck traffic. Nonetheless, a small

decrease of nitrogen oxide could be observed. This may indicate that urban heating is the major contributor to the Chamonix Valley pollution during winter.

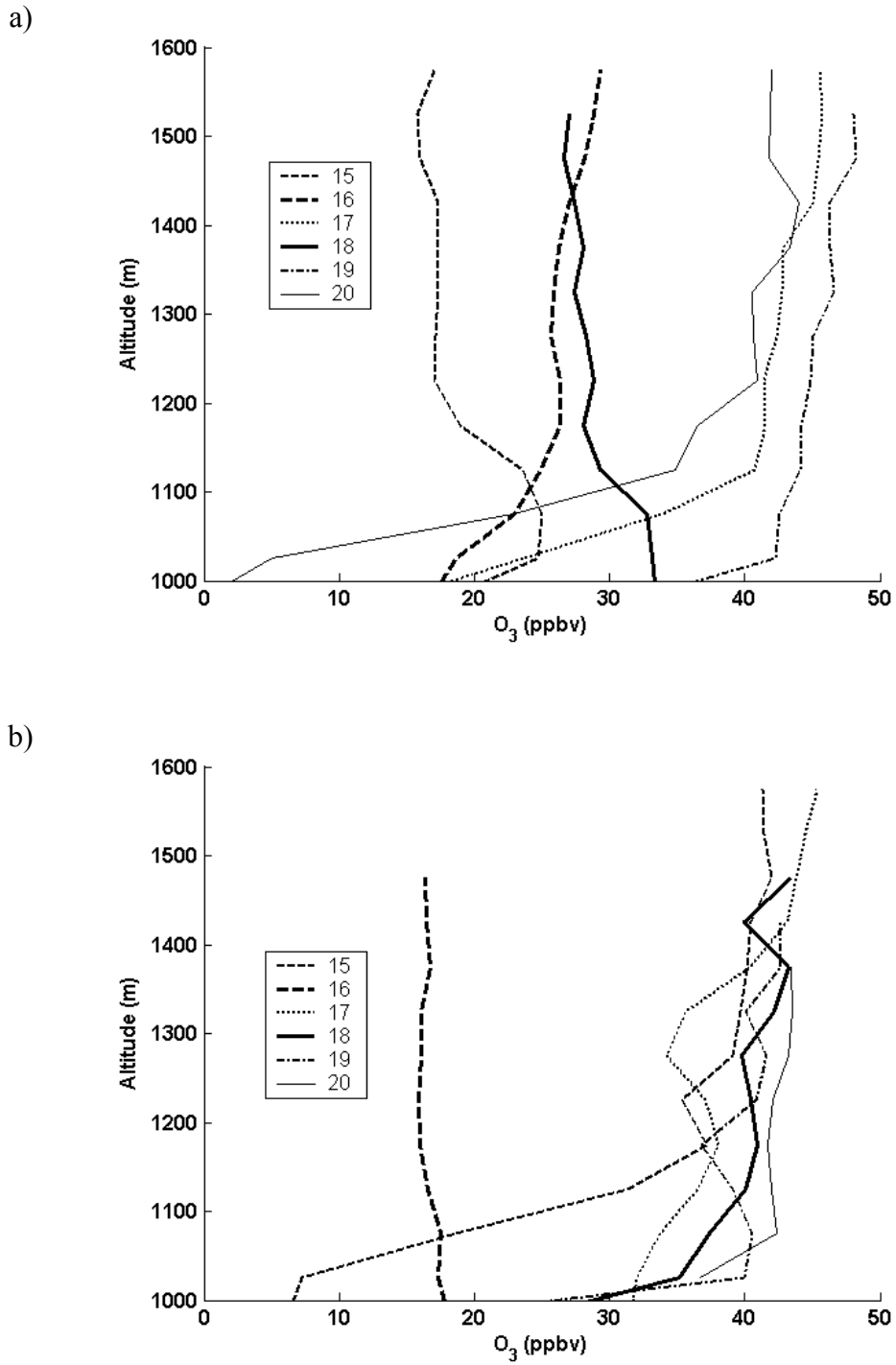
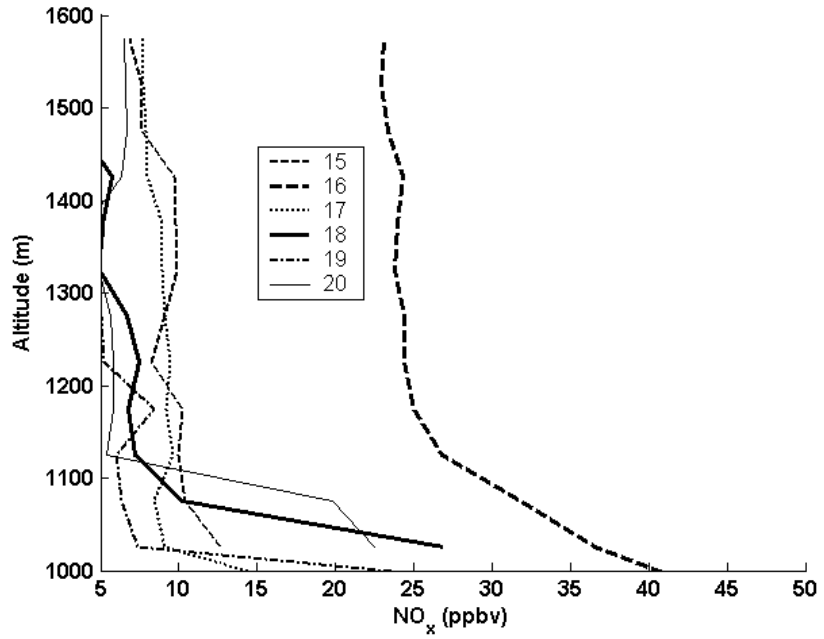


Figure 8: Same as Figure 4 for O<sub>3</sub>.

a)



b)

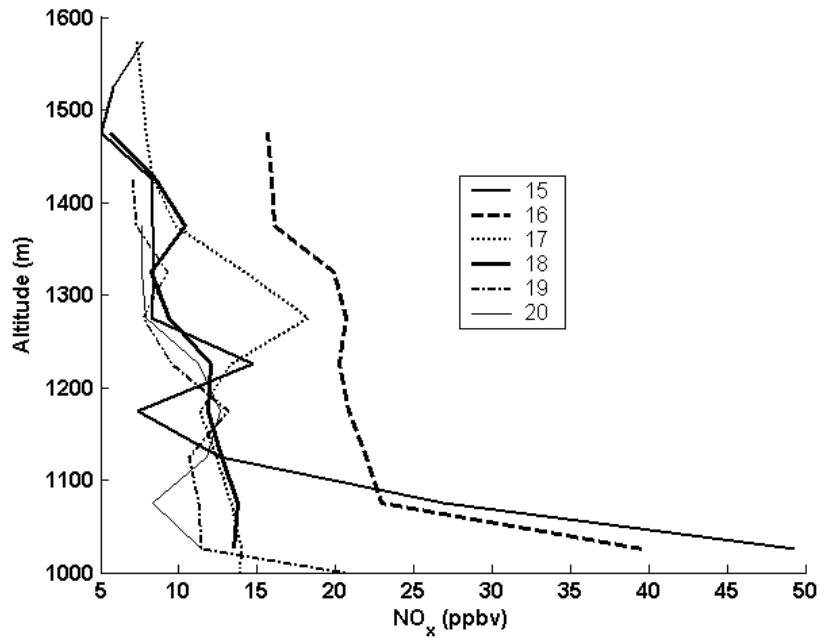


Figure 9: Same as Figure 4 for NO<sub>x</sub>.



#### **4. Conclusion**

It is the first time that measurements of mountain valley pollution are performed from ULA flights. It demonstrates the usefulness of this type of approach. Performed during winter in the Chamonix Valley, the measurements were obtained in the framework of the French program POVA. They have highlighted the strong vertical stability which persists in the valley throughout the day and stress the importance of local influences. This stability induces a trapping of the pollutants ( $O_3$ , NO,  $NO_x$  and aerosols) in the first 150 m avb and lingering pollution seems then to play a major role. An urban heat island effect has been well identified above Chamonix city with a vertical exchange of pollutants. Moreover, vertical profiles provide evidence for a persistent stable layer above the PBL. It could be considered as an injection layer where pollutants transit from the PBL to the free troposphere.

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## Notes des Activités Instrumentales

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