P3.6 COMPARISON OF POLLUTION EVENTS DURING THE ESCOMPTE 2001 CAMPAIGN : SEA-BREEZE AND SYNOPTIC WIND CONDITIONS

Aurore Brut[1], Frédérique Saïd[*1], Clotilde Moppert[1], Vincent Puygrenier[1], Bernard Campistron[1] and Pascal Perros[2]
[1]Laboratoire d'Aérologie, UMR CNRS 5560, Université Paul Sabatier, Toulouse, France

1. INTRODUCTION

The ESCOMPTE experiment was dedicated to the study of pollution events. A brief description of this program is presented in the companion paper (Moppert et al.) which is devoted to the description of the thermodynamic environment including the vertical structure of the low troposphere, during an intensive observation period (IOP2). A few results are first recalled here. This latter paper focuses on the difference between the first two days (2001 June 22 and 23: IOP2a) when the Mistral mesoscale wind is blowing and the three following days (2001 June 24 to 26 : IOP2b) when sea-breeze conditions and a weak wind are encountered. The boundary layer (BL) development is typical of a sunny summer day on June 22 only, reaching heights (zi) of 1900m in the early afternoon. The advection of colder air from sea limits the BL development that hardly reaches 1000m during IOP2b. A mesoscale effect due to the high pressure situation induces downward mesoscale vertical velocities that stem the BL growth. On June 23, a negative mesoscale velocity, not explained yet, also prevents the BL from growing until 15 UTC. One location in the ESCOMPTE area does not follow these general features : it is the entrance of the Durance valley, to the North-East of the domain, where the valley wind added to the Ventury effect enhances high development of BL (up to 2500m). To study the possibilities of transfers of pollution between the BL and the staggered layers capping the BL, a preliminary analysis is made using June 24 ozone profiles, as tracers of the various air masses. Except for the case of the Durance valley, the transfers are, most of the time, limited to a few number of cases and to a small depth in the low troposphere, due to the weakness of the BL developments.

The present paper is a preliminary work, based on experimental results, aimed at presenting the chemical conditions during IOP2, highlighting differences between IOP2a and 2b and checking whether the results pointed out for dynamics may alter chemistry.

[*]corresponding author address: Frédérique Saïd, Centre de Recherches Atmosphériques, 8 route de Lannemezan, 65300 Campistrous, France;
e-mail: saif@aero.obs-mip.fr

2. MEASUREMENTS

Radiosoundings or aircraft soundings are used as well as the horizontal fields obtained from aircraft parallel legs at 800m asl (except the northern leg above the Lubéron mountain that was flown at 1000m asl). The aircraft sensors are described and compared in Saïd et al. (2004). Three aircraft were involved: the Fokker 27 ARAT from the Institut National des Sciences de l'Univers, the Merlin from Meteo-France and the Dornier from the Institut für Meteorologie und Klimaforschung. The available data are the following : O_3 and CO on the 3 airplanes (1pt/s), NO and NO_2 on Dornier and ARAT (1pt/30s), fine particles number on ARAT (1pt/s), black carbon on ARAT (1pt/60s), CO_2 on Dornier (1pt/s). Radiosoundings (in Aix) also provided ozone measurements.

The flight strategy was to fly over the area three times per day to characterize the previous night conditions (5-7:30UTC), the best photo-dissociation conditions near midday (10:30-13UTC) or the accumulation/ transport result in the early afternoon (14:16UTC). Usually (except on June 22, when it flew at 10:30UTC), the DORNIER performed the afternoon flight. It is important to notice that NO measurements from DORNIER have to be taken with caution: NO_2 is obtained from a chemiluminescence reaction with Luminol, which is well controlled. NOx are deduced from a CrO_3 converter and NO is the difference between NO_x and NO_2. The efficiency of the conversion with CrO_3 is less trustful.

Turbulence was also measured and was very helpful to detect whether the 800m flight was performed inside or above the BL. The dissipation coefficient of turbulent kinetic energy (ε) was used to do so, as it is shown on Fig. 1. A black line divides the area in two parts with high value of ε inside the BL and low values outside.

3. JUNE 23 AND JUNE 26 DESCRIPTIONS

June 23 and June 26 were selected as an example for IOP2a and IOP2b but the same study was achieved for the whole IOP2. Results are shown in Fig. 2 and 3. Mistral conditions are best represented on June22 since the ARAT aircraft did not fly on this day, some fields are lacking. Only O_3, CO, NO and

NO_2 fields are displayed below for the early morning, midday and mid-afternoon. The same color scale (specific to each hour) was used on purpose to highlight the difference between both days. The thin black parallel lines are the tracks of the aircraft flights.

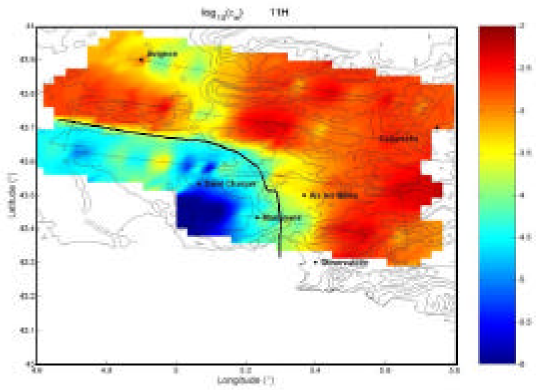

***Fig. 1** : Horizontal field of the dissipation coefficient of turbulent kinetic energy (e) measured at 11UTC on June 26. The black line delineates the area inside the BL (red and yellow) from the outside (blue).*

The first feature to note, which is not a surprise, is the difference between both days in the pollution level of the O_3 and CO fields : O3 and CO do not exceed 65ppb and 120ppb respectively on the Mistral event (June 23), whatever the day hour, whereas they reach 120 ppb and 320 ppb during the sea-breeze day at 11UTC. First, we can explain it by the stronger wind conditions of IOP2a which favour transport while the larger heights of BL favour dilution. However, we must keep in mind that the BL height was not so high on June 23 near midday, in the western part of the ESCOMPTE area. This argument of transportation and dilution is also hard to keep when looking at the NO and NO_2 fields : they exhibit well identified spots, that have been advected from the source areas (for instance the south-western yellow spot, which is over the sea, is coming from Fos, an industrial complex on the seashore), but that remain high, in comparison to the ozone concentration (67 ppb of O_3 and 2.5 ppb of NO_2 for the previous spot near Fos). The areas of high NO_2 concentration are even more polluted at 14UTC and spread on a larger area.

On June 26 at 11UTC, the ozone plume roughly follows the location of the highways and mean roads on the western and central part of the domain (Fig. 1 in the companion paper). These traffic areas cross anyhow the industrial and urbanized areas. In the afternoon, sources of CO, NO and NO_2 are located in the same areas. However these fields do not allow to study the south-western part, due to the weakness of z_i : we have no means of knowing whether the plume also reaches this area. In the afternoon, the ozone plume has been transported to the North-East (south sea-breeze associated to synoptic south-western wind). CO and NO primary pollutants have decreased but the NO_2 concentration has increased (titration reaction).

An interesting feature, in relationship with the dynamics highlighted in the companion paper can be pointed out for the area South to Cadarache, at 5UTC on June 26. Some primary and secondary pollutants have accumulated during the night (CO, NO and NO2, O_3). Due to the more efficient development of the BL in the north-eastern part of the domain, this pollution spot has vanished at 11UTC.

The vertical explorations (not shown here) enabled to study the sublayers capping the BL. On June 23, the ozone concentration of the first sublayer (starting at 1000m) was systematically 25ppb less than that of the BL. NO2 grew from 0.4 ppb (5UTC) to 1.3 ppb (15:30UTC) while NO remained at 0.05 ppb. The dynamic study indicated that the development of the BL occurred rather late on this day : consequently the concentration of pollution of the afternoon flight may have been decreased due to dilution after 14:30UTC.

It is explained in the companion paper that a possible mixing is less likely to occur during IOP2b, due to the weak development of BL. Anyhow, in this case, even if a mixing occurs, it will either let the BL concentration unchanged or it will increase it since the capping sublayers are sometimes more polluted than the BL. This confirms the well-known fact that severe pollution episodes often occur in a context of pollution at large-scale, such as the scale of the continent.

4. RESULTS FOR THE WHOLE IOP

We tried to sum up the results of the whole IOP by extracting information of both vertical soundings and horizontal fields. On both representations, homogeneous areas of a parameter like O_3 were identified, or on the contrary, points with maximal or minimal values were selected. The corresponding value for other chemical parameters was also noted. The result is plotted on Fig. 4 in terms of BL measurements or measurements in the upper layers. The relationships shown in this figure cannot be considered as a statistical view of the IOP2, however it is a good indication of the most striking features. The following points can be pointed out from the curves :

- ozone concentrations are larger in IOP2b (60-130ppb) than in 2a (40-60ppb). Midday and afternoon concentrations are similar within

each IOP, except for 2 large values of IOP2b (June 26) : after midday, O_3 is transported but not produced anymore.
- $N0_x$ are always weaker over zi (around 0.05 ppb or 0.01ppb for NO2 and NO respectively), whatever the time and the IOP, which is not surprising since NO_x are not generated in the upper layers. However, the range of NO_2 above zi is larger during the afternoons of IOP2b (1 to 2 ppb), indicating that more polluted air masses were transported from other areas with higher zi. The relationship between O_3 and CO also shows higher concentrations of CO outside the BL during IOP2b (similar to those of the BL during IOP2a).
- NO_2 concentrations in the BL are the same, whatever the time (0.05-4ppb). The striking point is that this range does not vary with the IOP : NO_2 concentrations are as important in IOP2a as in IOP2b whereas transport and dilution due to higher BL should have reduce their concentration. Moreover, O_3 is not generated : the same concentration of NO_2 does not yield the same concentration of O_3. There are probably several explanations for these mechanims : in spite of the sea-breeze, mean temperatures were higher during the anticyclonic conditions of IOP2b which enhances ozone production. Furthermore, the moistening of the area due to the synoptic winds (West or South) favors reactions with hydrocarbons and produces O_3.
- the case of NO is similar : some large NO values occur in IOP2a as in 2b for lower O_3 values in IOP2a. The very large values of NO in IOP2b correspond to the largest ones of O_3 and NO2, previously pointed out for the 26.
- CO is very high and not connected with NO_x in the middle of the day (no clear relationship), whereas CO and O_3 grow together with a logaritmic shape. In the afternoon, the relationship turns almost linear for O_3 and CO, and NO_2 and CO start to get connected. CO, NO_2 and O_3 have been transported away from the sources during the afternoon, they may evolve similarly.

5. CONCLUSION

This study focusing on chemistry processes during the ESCOMPTE second IOP raises more questions than it brings answers. If the level of ozone pollution is less important during the Mistral days, primary and secondary pollutants like NO, CO and NO_2 keep very high values, so that transport or dilution cannot be used to explain the low levels of ozone. However, transport may not be totally inefficient : most of early morning horizontal fields of IOP2b show that primary as well as secondary pollutants accumulated during the night, in the case of the light wind conditions of IOP2b. Meanwhile, temperature and moisture conditions remained high, which favoured ozone production.

Another important result that was brought by the study about dynamics is that BL heights are not very high during IOP2b: moreover, if ever they turn to be deep enough to reach the upper layers, this leads to an increase of ozone concentration, due to the level of pollution at large scale, advected from remote areas. On the contrary, the air capping the boundary layers of Mistral days is cleaner.

6. BIBLIOGRAPHY

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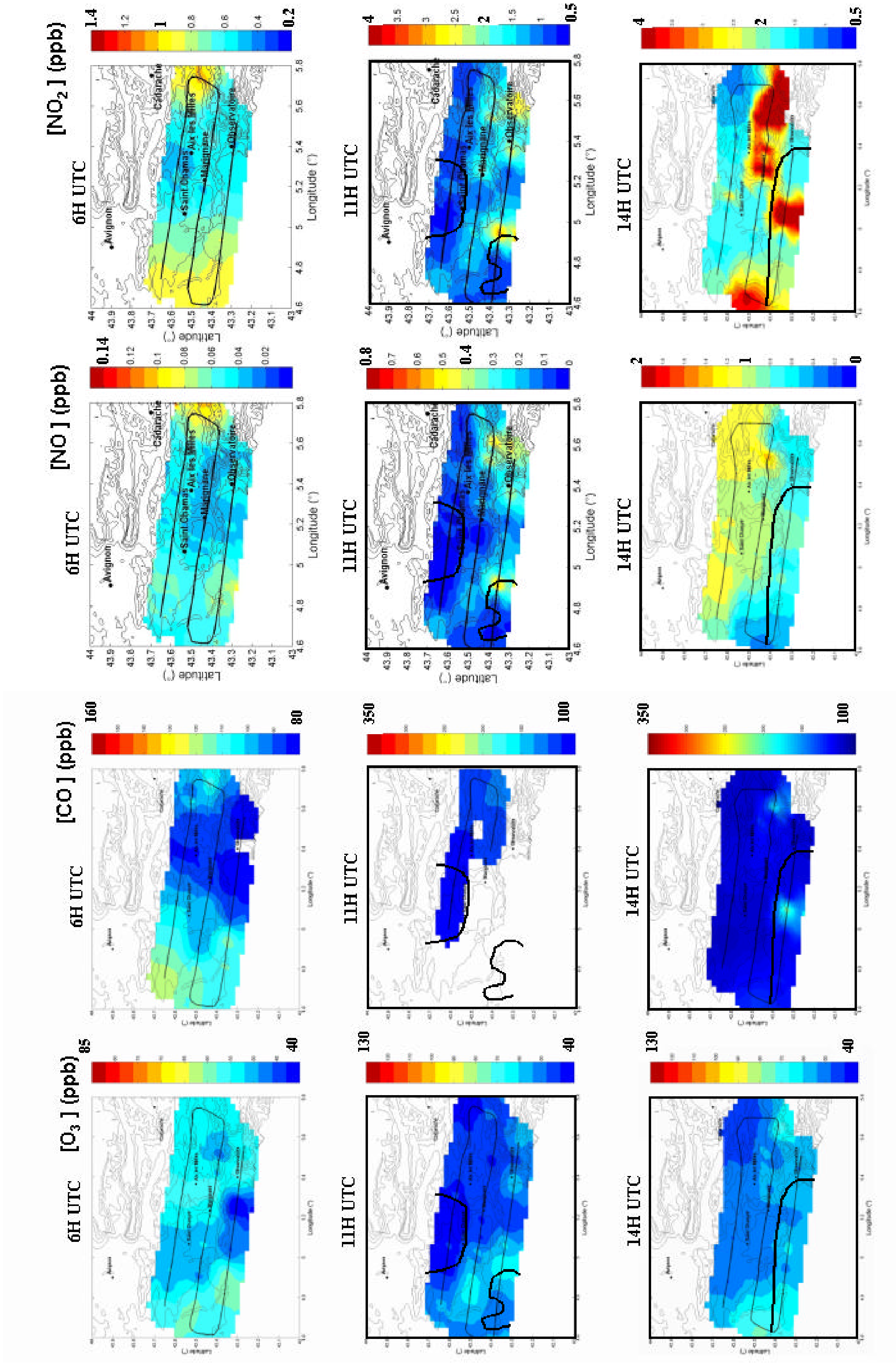

***Fig. 2 :** June 23 horizontal fields of O3, CO, NO and NO2 (from left to right) starting at 5, 11 or 14 UTC (from top to bottom). The thin black line represents the flight track, the solid one is explained in the text.*

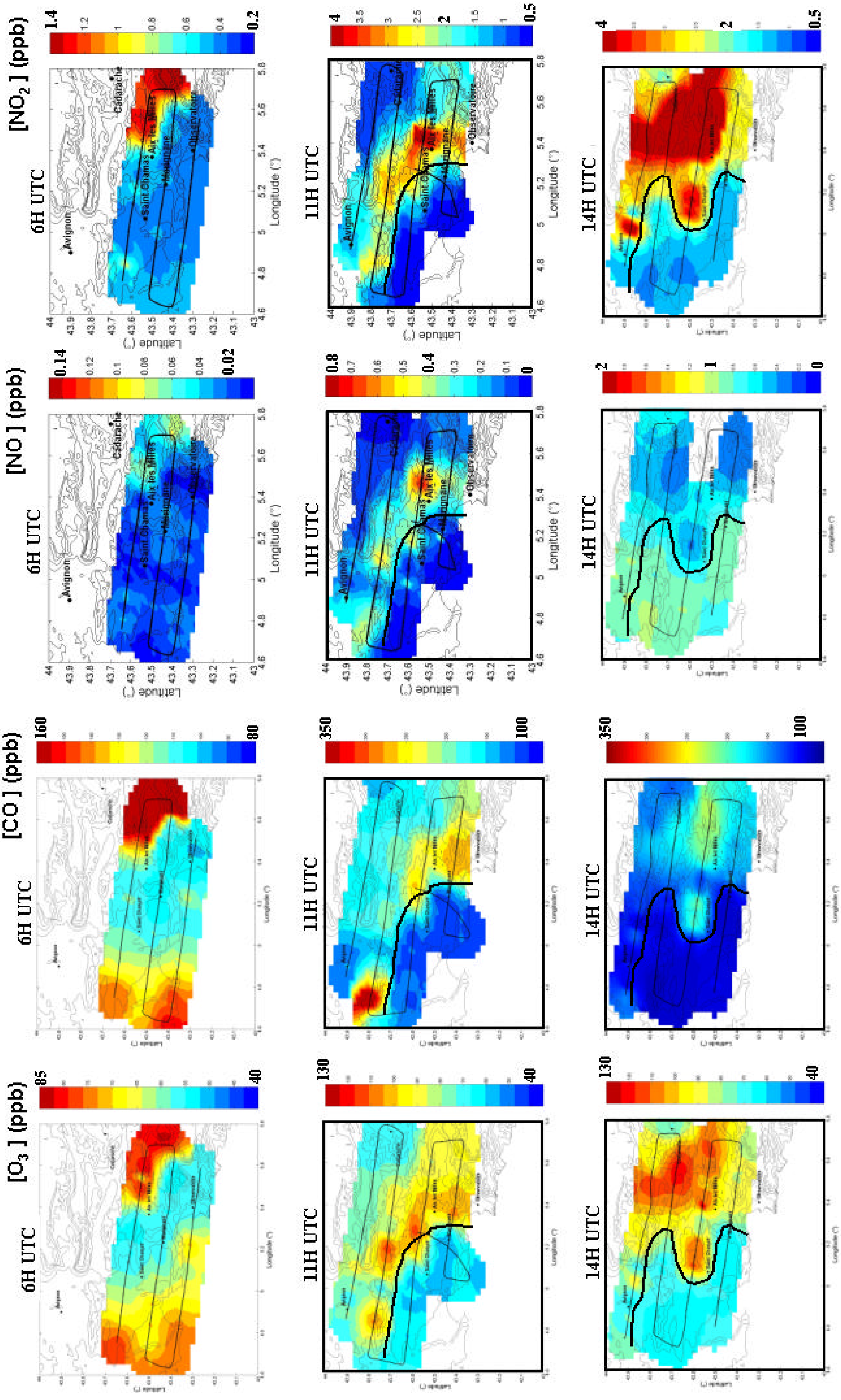

***Fig. 3 :** Same as Fig. 2 for June 26.*

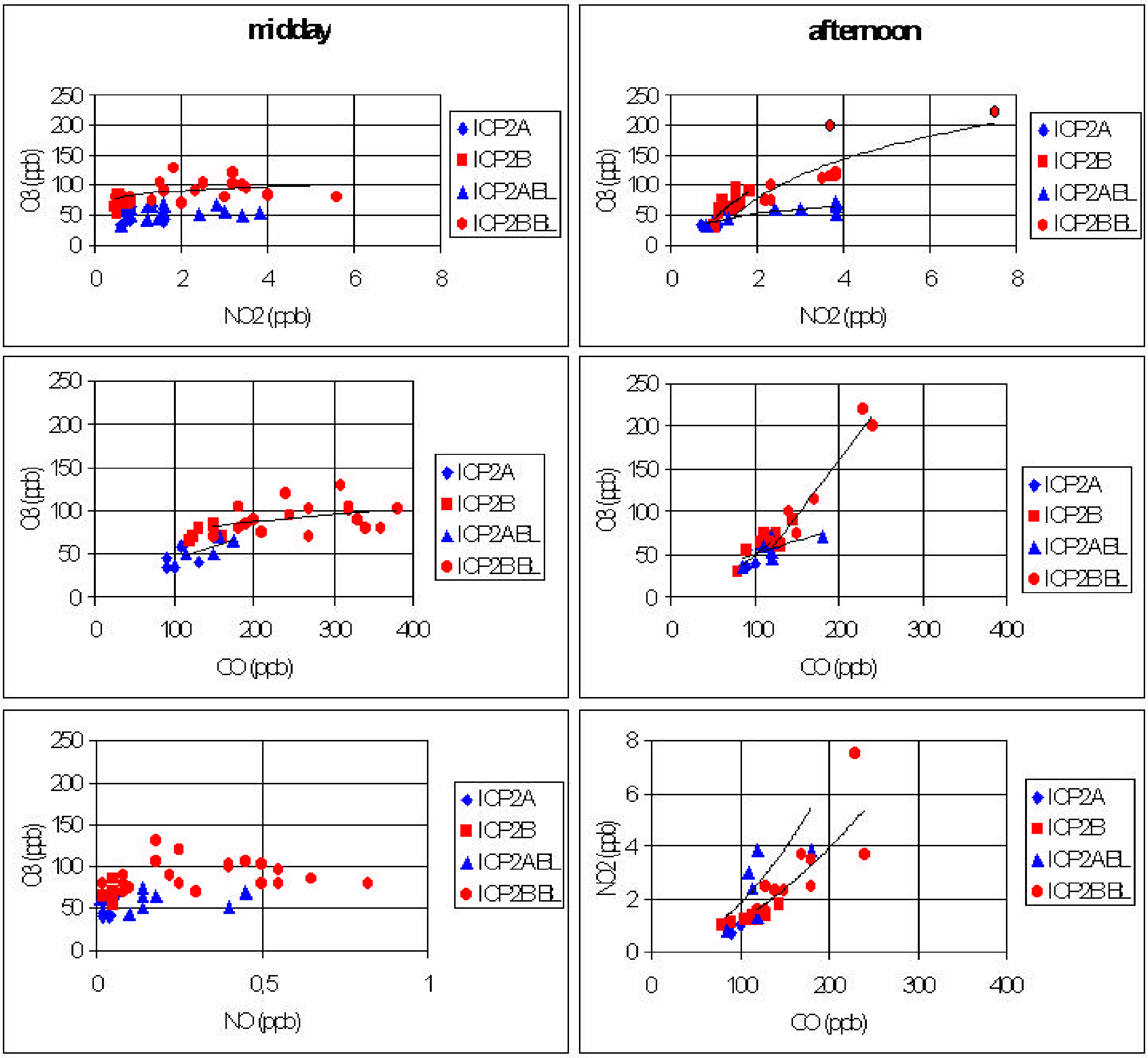

***Fig. 4 :** Midday and afternoon relationships between ozone, N0$_x$ and CO during IOP2a (blue) and IOP2b (red) for boundary layer measurements (refered as 'BL') and measurements in the upper layers.*