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# HIGH RESOLUTION AIR MODELING IN A DEEP VALLEY: ANALYSE OF CHEMICAL INDICATORS FOR MANGEMENT OF ROAD TRAFFIC

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#### ABSTRACT

Road traffic is a serious problem in the Chamonix valley: road traffic and air pollution worry the inhabitants. The big fire in the Mont-Blanc tunnel made it possible, in the framework of the POVA project (Alpine Valley Pollution), to undertake measurement campaigns with and without heavy-vehicle traffic through the valley, towards Italy (before and after the tunnel re-opening).

Atmospheric prediction model ARPS 4.5.2 (Advanced Regional Prediction System), developed at the CAPS (Center for Analysis and Prediction of Storms) of the University of Oklahoma, enables to resolve the atmosphere dynamics above a complex terrain. This model is coupled to the TAPOM 1.5.2 atmospheric chemistry (Transport and Air POllution Model) code developed at the Air and Soil Pollution Laboratory of the Ecole Polytechnique Fédérale de Lausanne. The numerical codes MM5 and CHIMERE are used to compute large scale boundary forcing. Using 300-metre grid cells to calculate the dynamics and the reactive chemistry makes possible to accurately represent the dynamics in the valley (slope and valley winds) and to process chemistry at fine scale.

Atmospheric chemistry indicators are computed for days of the field campaign. NOy,  $H_2O_2/HNO_3$ ,  $O_3/NOz$  demonstrate a VOC controlled regime. A new indicator based on ozone chemistry contribution and computed from the chemistry code is suggested for the purpose of localising ozone production area in the valley. Various scenarios of traffic reduction demonstrate a significant increase of ozone concentration at night.

#### **INTRODUCTION**

Alpine valleys are sensitive to air pollution due to emission sources (traffic, industries, individual heating), morphology (narrow valley surrounded by high ridge), and local meteorology (temperature inversions and slope winds). Such situations are rarely investigated with specific research programs taking into account detailed gas atmospheric chemistry.

Following the accident under the Mont Blanc tunnel (Fig.1) on 24 March 1999, international traffic between France and Italy was stopped through the Chamonix Valley (France). The heavy-duty traffic (about 2130 trucks per day) has been diverted to the Maurienne Valley, with up to 4250 trucks per day. The POVA program (POllution des Vallées Alpines) was launched in 2000.

The general topics of the program are the comparative studies of air quality and the modeling of atmospheric emissions and transport in these two French alpine valleys before and after the reopening of the tunnel to heavy duty traffic in order to identify the sources and characterize pollutant dispersion. The program includes several field campaigns, associated with 3D modeling in order to study impact of traffic and local development scenarios.



Fig. 1. Topography of Chamonix valley: main measurement sites and coordinate system UTM 32 in meters.

# MODEL FOR SIMULATION

The Chamonix valley (23 km long) is a deep: 1km wide at the bottom and 5 km from ridge to ridge with altitude ranging from 1000 to 4810 meters. We use a modeling system: numerical simulations combine the meso-scale atmosphere model ARPS 4.5.2 and the troposphere chemistry model TAPOM 1.5.2.

# Model for atmosphere dynamic

Large eddy simulation was used to compute meso-scale flow fields. The numerical simulations presented here have been conducted with the Advanced Regional Prediction System (ARPS), version 4.5.2 ([1], [2]) with a horizontal resolution of 300 meters. Lateral boundaries conditions were externally-forced from the output of larger-scale simulations performed with the Fifth-Generation Penn State/ NCAR Mesoscale Model (MM5 v.3) ([3]).

# Model for atmosphere chemistry

ARPS is coupled off-line with the TAPOM 1.5.2 code of atmospheric chemistry (Transport and Air POllution Model) developed at the LPAS of the EPFLausanne ([4], [5]). 300-meter grid cells to calculate dynamics and reactive chemistry make possible to accurately represent dynamics in the valley (valley and slope winds) ([6]) and to process chemistry at fine scale.

TAPOM is a three dimensional eulerian model with terrain following mesh using the finite volume discretisation. It includes modules for transport, gaseous and aerosols chemistry, dry deposition and solar radiation.

TAPOM uses the Regional Atmospheric Chemistry Modeling (RACM) scheme ([7]). The mechanism includes 17 stable inorganic species, 4 inorganic intermediates, 32 stable organic species (four of these are primarily from biogenic origin) and 24 organic intermediates, in 237 reactions. In RACM, the VOCs are aggregated into 16 anthropogenic and three biogenic species.

For the boundary conditions, CHIMERE, a regional ozone prediction model, from the Institut Pierre Simon Laplace, gives concentrations of chemical species at five altitude levels ([8]) using its recent multi-scale nested version.

# **EMISSION INVENTORY**

The emission inventory is based on the CORINAIR methodology and SNAPS's codes, with a 100x100m grid and includes information (land use, population, traffic, industries...) gathered from administrations and field investigations. The emission inventories are space and time-resolved and include the emissions of NOx, CO, CH4, SO2 and volatile organic compounds (VOC). Biogenic emissions from forests and grassland are included. All the emissions are lumped into 19 classes of VOC as required for the Regional Atmospheric Chemistry Mechanism (RACM) ([7]).

Emission classes are given in the table 1. The emission inventory takes into account roads and access ramps to the tunnel adjusting emissions to the slope of the road. A specific feature of emission is the significant contribution of heavy vehicles (> 32 tons).

Traffic sources	Anthropogenic sources	<b>Biogenic sources</b>
Heavy vehicles	Commercial boiler	Forest
Utilitarian vehicles on motorway	Residential boiler	Grassland
Utilitarian vehicles on road	Domestic solvent	
Cars	Gas station	
Cars in city		-
Aerial traffic		

Table 1: Classes of the emission inventory.

# VALIDATION

A first step in modeling Chamonix valley was to compute a simplified case with no synoptic forcing and with open boundary conditions ([9]). Then, atmospheric circulations develop by themselves from thermal processes only. Such a configuration enhances features specific to the valley and mimic the worst conditions for pollution because of a lack of mean transport. This idealized case is not so far from the situation frequently observed in Chamonix with dynamics inside the valley decoupled from synoptic meteorology. Thus dynamics is restricted to convection and slope wind from sun heating (solar radiation was chosen to correspond to June).

The simulations presented here are not realized in this simplified case but take a full account of the real meteorology of the week of computation during the summer IPO (05/07/03 to 11/07/03).

More details on dynamics and on chemistry validation and process will be described in separate papers.

# High-resolution meteorological simulation.

The redistribution of pollutants and therefore the ozone production is very dependent on meteorological conditions. The observed meteorological situation during 7 days of intensive period of observation (IPO) is summarized in table 2. A northwesterly wind with sun prevailed. In this complex mountainous area, wind balance and slopes wind are important for the transport of chemical species.

	05/07/03	06/07/03	07/07/03	08/07/03	09/07/03	10/07/03	11/07/03
Description of the situation	₩¥	₩¥	A A	A.	and the second s	A.	
Tmin (°C)	4	5	6.5	7	8.5	8	8
Tmax (°C)	22	24	25	26	26	28	27
Isotherm 0°C	3700 m	3850 m	3700 m	4200 m	4000 m	4100 m	4000 m
Wind	NW	NW	W	(<1m/s)No	(<1m/s)No	N to NW	NW
description at	2.5 m/s	4 m/s	4 m/s	significant	significant	5.5 m/s	7 m/s
4500 m asl							

Table 2: IPO meteorology.

Wind Profiler data are in good agreement with values from the model (fig. 2): wind balance starts and stops at the same time. The altitude of the synoptic wind is well represented. Discrepancies are observed on July,  $9^{th}$  but it is a stormy day with instable weather. The

boundary layer thickness is well simulated all along the day as it may be observed from the vertical potential temperature profile.



Fig. 2. Wind force and wind direction from the wind profiler (left-hand side) compared to results from the computation (right-hand side).

Simulated meteorological fields are realistic: the evolution of the height of the inversion layer is well simulated. Wind direction and force is well reproduced with wind reversal observed at the same times in the model and from measurements. Therefore, meteorological fields may be viewed as realistic enough to drive transport and mixing of chemical species.

#### High-resolution chemistry simulation.

Concentrations of pollutants in the valley (such as  $O_3$  or  $NO_2$ ) are rather low at least when compared with large cities: values peak at 75 ppb for  $O_3$  and 40 ppb for  $NO_2$  compared to 100 ppb for  $O_3$  and 50 ppb for  $NO_2$  in nearby city of Lyon or Grenoble.

Ozone from the observation and from the model is in good agreement in both urban and rural station. Both spatial and temporal variability of the simulated ozone concentrations correspond reasonably well to the measured values. Fig. 3 shows the correlations between the measured and simulated ozone concentrations for the whole IPO week except for the stormy day (9 of July). Coefficients show high values with  $0.73 < R^2 < 0.76$ .



Fig. 3. Comparison between measured and simulated ozone in three sites for the IPO.

These results can be compared to ozone correlations in Grenoble during a high ozone episode with  $R^2=0.64$  for urban station and  $R^2=0.42$  for suburban stations ([10]). Background station ('Col des Montets' and 'Plan de l'aiguille') are directly under regional influence. The amplitude of the variation of ozone concentrations are low, therefore it does not make sense to give correlation coefficient. The relative mean error on ozone concentration all along the IPO (with the stormy day included) is 12% at the site 'Plan de l'aiguille' and 16% for the site 'Col des Montets'.

# PHOTOCHEMICAL INDICATORS TO CHARACTERIZE OZONE PRODUCTION REGIME

Narrow valleys in mountainous environment are very specific areas when it comes to air quality. Emission sources are generally concentrated close to the valley floor, and very often include industries and transport infrastructures. In order to develop ozone abatement strategies it is important to know whether in a specific area the ozone production is limited by VOC or NOx. In order to understand the impact of the emissions sources on ozone production regime, three simulations were performed. All of them are based on meteorology and emission inventory of July 6<sup>th</sup> 2003. Run B is the simulation of July 6. Run N corresponds to an arbitrary reduction in NOx emissions of 50%. Run V is obtained with an arbitrary reduction in VOC emissions of 50%. The three runs are described in table 3.

	Date	Duration	Emissions
Run B	6 July 2003	24 hours	All
Run N	6 July 2003	24 hours	Run B – 50% NOx
Run V	6 July 2003	24 hours	Run B – 50% VOC

Table 3: Runs to determine ozone production regimes

6 July 2003, is representative of a summer sunny day with mean pollution level. Photochemical indicators are considered in order to make the difference between NOx limited and VOC limited ozone formation.

The indicators under consideration are: NOy (NOy=NOx+HNO<sub>3</sub>+PAN) ([11]), O<sub>3</sub>/NOz (NOz=NOy-NOx) and H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> ([12]). These indicators have been tested in many modeling studies and for different places like Mt. Cimone, the highest mountain in the northern Italian Apennines ([13]), like Paris ([14]), or like Grenoble ([10]).

Fig. 4 illustrates the NOx-VOC sensitivity for the simulations (runs N and V) over the bottom of the valley. Only meshes of the terrain at an altitude less than 1500 meters above see level are considered in order to include all anthropogenic sources. Although a significant part of the domain area is rural-type, effects of non rural emission predominate.



Fig. 4.  $H_2O_2/HNO_3$ ,  $O_3/NOz$ , NOy indicators and  $\Delta O_3$  difference of ozone concentration with reduction of VOC or NOx, on 6 July 2003 in the bottom of the valley.

The figure 4 shows changes in ozone concentrations associated with either reduction of VOC (run V) or reduction of NOx (run N) relative to the domain. The positive values represent locations where, by decreasing the emission, a reduction in ozone is obtained while negative values result from locations where reduction of emissions produces more ozone. According to the results with these three indicators, the ozone production is VOC limited: only a diminution of VOC leads to a reduction of ozone concentration (run V). This conclusion differs from what was observed in the nearby city of Grenoble (100 km from the valley in a Y shape convergence of three deep valleys) where a NOx controlled regime was observed ([10]).

But it is generally thought that in the densely populated regions of northern Europe, ozone is more likely to be sensitive to emissions of VOC than to NOx. The situation in northern Europe stands in contrast with results from southern Europe and parts of the eastern U.S., where ozone is often sensitive to NOx rather than to VOC and where biogenics have a significant impact ([15],[12]).

# HYBRID INDICATOR TO DETERMINE OZONE PRODUCTION AREAS

Ozone production sites can be different of sites with high ozone concentrations: it is a secondary pollutant submitted to transport.

For the purpose of defining strategies to improve air quality, the observation of times and locations when and where ozone is produced would be more useful than probing concentrations. For instance, significant production of ozone may be associated to relatively moderate ozone concentration inside area under constant wind. Contrary to field observations, numerical models allow to compute rates of production for every chemical species.

TAPOM is based on the resolution of mass balance equation for concentration of 77chemical species. The concentration of ozone results from contributions from wind transport, turbulent diffusion, dry deposition, emissions and chemical contribution (1). Emission does not contribute to concentration of secondary species.

(1) 
$$\frac{\partial [O_3](x_i,t)}{\partial t} + \overline{U}_i \frac{\partial [O_3]}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \frac{\partial (K_{ij}[O_3])}{\partial x_j} \right) - \left( \frac{v_d}{dx_k} \right) \frac{\partial [O_3]}{\partial x_i} + W_{O_3}(x_i,t)$$

With U the mean speed, K the diffusion turbulent coefficient,  $V_d$  the speed of deposition and W the chemical reaction rate.

The net contribution of chemistry to ozone production is:

(2) 
$$W_{O_3} = P_{O_3} - L_{O_3} C_{O_3}$$

With P the production rate and LC the consumption rate of ozone.

The chemical reaction rate (W) from chemistry (production-consumption) is explicitly computed at each time step t and in each location  $x_i$  for every species, including ozone. Therefore, the contribution from photochemistry can be identified with its distribution in space and time at the resolution of the computational grid.

The chemical reaction rate (W) includes both production and consumption steps, therefore W may be either positive or negative.

Ø is defined as:

High positive values of Ø stand for locations of ozone net production, and negative values for ozone net consumption.

Output of W and ozone concentration is done every hour in each node of the simulation domain. The indicator  $\emptyset$  on 7 July 07:00 LT shows relative important values (-1.5< $\emptyset$ <-4) located at the entrance of the valley, all along the motorway to the tunnel entrance. These high values may be partly attributed to weak concentration of ozone in this location (30< [O<sub>3</sub>] <40). Ozone is consumed because of VOC controlled regime.





O3 augmentation (ppb) - wighway and tunnel - 07/07/2003

Fig 5. Value of Ø on the motorway, 7 July 2003.

Fig. 6.  $O_3$  augmentation with scenario of reduction traffic.

The indicator  $\emptyset$  shows high and negative values along the motorway between 06:00 and 09:00 LT (fig. 5). All along the day,  $\emptyset$  is negative and around -1: there is more consumed ozone than produced ozone but with a mix due to transport.

Indicator  $\emptyset$  shows high and negative value in the area of the motorway, because of the traffic. Traffic emissions are made of 5.7 tonnes of NOx and 1.2 tonnes of non methanic VOC for the day of the simulation. This area as being not so densely populated is not under significant influence of residential emission.

Then, we compute a new simulation without the road traffic in the valley, between 06:00 and 09:00 LT. We can observe an increase of ozone concentration: 5 to 15 ppb in the bottom of the valley in the highway area, and 45 ppb near the tunnel entrance (fig. 6). With this scenario, we observe an augmentation of  $\emptyset$  (fig. 5) with values around 0.

It is possible to conclude for ozone, that the stop of early morning road traffic leads to an important increase of concentration as prevue by the VOC controlled regime and $\emptyset$ .

# **TRAFFIC SCENARIOS**

Various scenarios are run in order to assess respective impact of heavy duty and light duty traffic with a unique meteorology. Four emission scenarios are performed during two validated days: 6 and 7 of July 2003 (table 4).

<b>S0</b>	Validated days: all realistic emissions
<b>S1</b>	no heavy duty traffic
<b>S2</b>	no light traffic
<b>S3</b>	50% of all road traffic

Table 4: scenarios performed.

We compare the 4 simulations at 3 measurements site in the bottom of the valley:

- Les Houches: in the valley entrance, near the tunnel.
- Chamonix: the biggest city of the valley, in his centre.
- Argentière: village at the opposite of the valley entrance.

Only simulations S2 and S3 lead to a significant change in the bottom of the valley for ozone,  $NO_2$  and CO concentrations (figure 7).

The VOC controlled regime of the valley leads to an increase of ozone with traffic reduction. These increases of ozone are more important with a reduction of all the traffic (S3) and of the light traffic (S2) and at night between 18:00 and 6:00 LT. The maximum of increase is 46 ppb at 22:00 LT, site 'Les Houches' for a reduction of 50% of all the traffic (S3). During day, concentration and maximum of ozone are similar for the three sites and all scenarios.

For  $NO_2$  and CO, concentrations highly decrease (the emitted volume of NOx and VOC decrease) for scenarios with traffic reduction. There is less than 10 ppb of  $NO_2$  against 50 ppb without reduction of traffic. CO concentrations shows a constant background around 100 ppb with scenarios of reduction of traffic, under peaks at 500 ppb without traffic reduction.

The reduction of heavy duty traffic (S1) does not change clearly ozone concentration in the valley but the intensive period of observation was a transitional period for this traffic. Indeed, after the reopening of the tunnel, drivers do not immediately change habitudes to pass through the Fréjus tunnel (Maurienne valley). Traffic flow for heavy duty vehicles, during the summer IPO, was half the classical traffic. In this study, with the various scenarios performed, ozone concentration is more sensitive at light traffic.



Fig. 7. Concentrations of O<sub>3</sub>, NO<sub>2</sub> and CO for 4 scenarios at sites 'Les Houches', 'Chamonix' and 'Argentière'.

#### CONCLUSIONS

Three-dimensional photochemical model simulations have been performed for 7 days, during the intensive period of observation in the topographically complex and narrow valley of Chamonix valley. Results from the numerical simulation are in good agreement with observations. Based on the model scenarios over Chamonix and on the combination of the indicators NOy,  $O_3/NOz$  and  $H_2O_2/HNO_3$  one can say that the region of the maximum ozone is VOC saturated.

The introduction of a new indicator ( $\emptyset$ ), defined as the ratio of hourly ozone production (from the numerical model) to the ozone concentration, allows to localise ozone production. Because of wind it is possible to find high ozone production with low ozone concentration. With the special VOC control regime of the valley,  $\emptyset$  demonstrates that a stop of traffic, between 7:00 and 9:00 LT on June 7, leads to an important ozone concentration augmentation (10 to 40 ppb).

Different scenarios for traffic emissions demonstrate (with the traffic of the IPO) an important sensibility of ozone concentration to the light traffic. Reduction of traffic leads to an augmentation of ozone concentration at night. Diminution of heavy duty traffic, with the model, results in a diminution of CO and NO<sub>2</sub>, but do not change clearly  $O_3$  concentration.

With the transfer of traffic from Chamonix to Maurienne valley because of the accident of Mont-Blanc tunnel, program POVA investigates also Maurienne. As for Chamonix valley, primary and secondary pollution is considered with measurements and numerical model. Ozone production regime and indicators obtained in the two valleys will be compared.

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