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Contribution of atmospheric processes to photochemical pollution by using a process analysis tool in the north-eastern and central Iberian Peninsula

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Background

- The Mediterranean and specifically the Iberian Peninsula (IP) undergo frequent photochemical pollution episodes during summertime, in which O_3 and PM10 exceedances of the European air quality targets are commonly registered (*Jiménez et al., 2006*).

- The knowledge and understanding of atmospheric processes leading to these pollutants levels and the weight of anthropogenic emission sources in their origin are fundamental in order to improve air quality.

- A number of studies have shown that during typical summertime conditions layering and accumulation of pollutants such as ozone and aerosols were taken place along the eastern coast of the Iberian Peninsula by means of experimental techniques, such as Lidar measurements (Baldasano et al., 1994; Pérez et al., 2004). Recently, Jiménez et al. (2006) used the ECHAM5/MESSy modelling system to perform a O_3 budget in the North Eastern Iberian Peninsula, taking into account the contributions of diffusion, advection and convection, wet and dry deposition, chemical production and stratosphere-troposphere exchange.

- The air quality models usually provide the net concentration of pollutants. In addition, some grid models can be configured to provide quantitative information on the effects of the chemical reactions and other atmospheric processes that are being simulated (*Jeffries and Tonnesen, 1994; Jang et al., 1995*).

- The process analysis is currently implemented in the Models-3 Community Multiscale Air Quality (CMAQ) modelling system, consisting basically in two modules: the Integrated Process Rate (IPR) and the Integrated Reaction Rate (IRR), which can be switched on separately (*Gipson, 1999*). The IPR provides the effects of all the physical processes and the net effect of chemistry on model predictions.

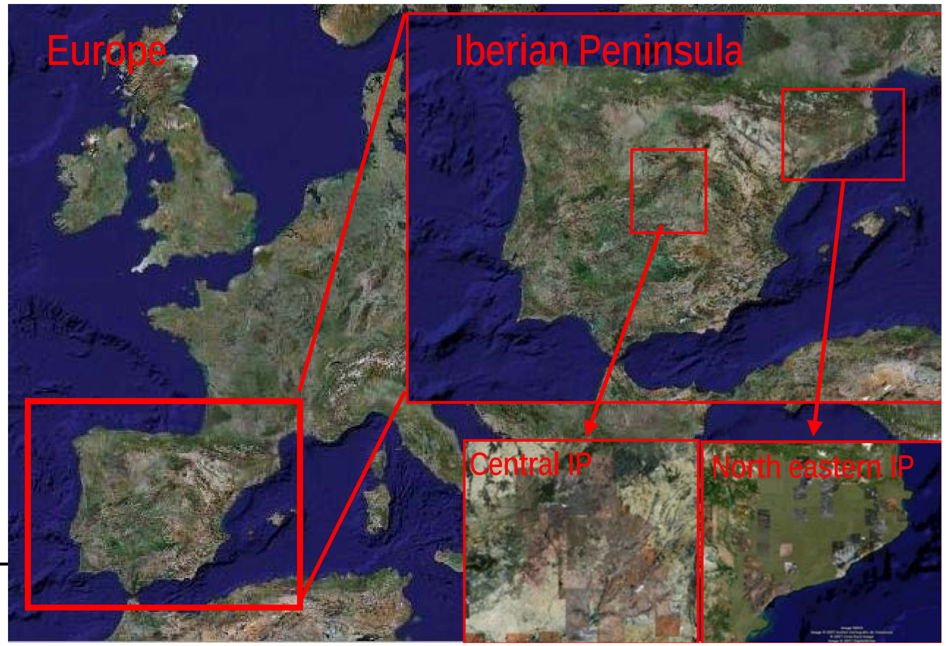
Objective

- This work quantifies the **contribution of different atmospheric processes to O_3 net concentration** in the **north-eastern and central Iberian Peninsula**, where major urban areas of Spain are located: Barcelona and Madrid, by using the **process analysis** tool available in the CMAQv4.6 model.
- **Advection and convection, horizontal and vertical diffusion, dry deposition, gas-phase chemistry, clouds interactions and wet deposition are considered.**
- The period of study is a photochemical pollution episode of 2004, the most recent year in which all necessary data were available: the **17-18, June.**

Available processes in the CMAQv4.6 Integrated Process Rate Analysis

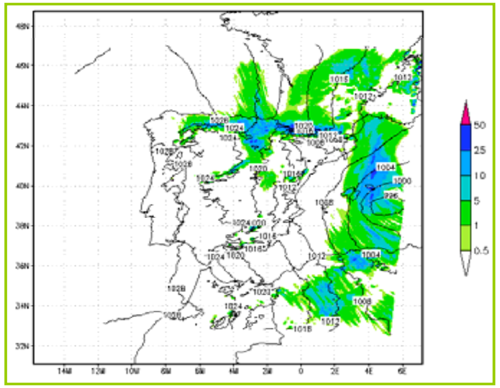
PACP Code	Process Description
XADV	Advection in the E-W direction
YADV	Advection in the N-S direction
ZADV	Vertical advection
ADJC	Mass adjustment for advection
HDIF	Horizontal diffusion
VDIF	Vertical diffusion
EMIS	Emissions
DDEP	Dry deposition
CHEM	Chemistry
AERO	Aerosols
CLDS	Cloud processes and aqueous chemistry
PING	Plume-in-grid

Location of the study areas



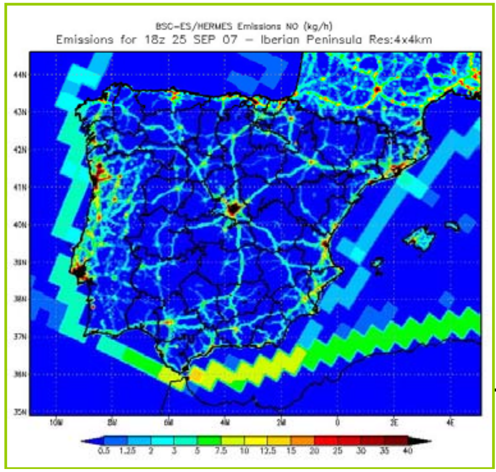
Methods: modelling system and configuration (1/2)

- The WRF-ARW/HERMES/CMAQ modelling system was applied to the north eastern and central Iberian Peninsula during the photochemical pollution episode of 17-18 June, 2004; with high resolution (1 $km^{2,}$ 1 h).

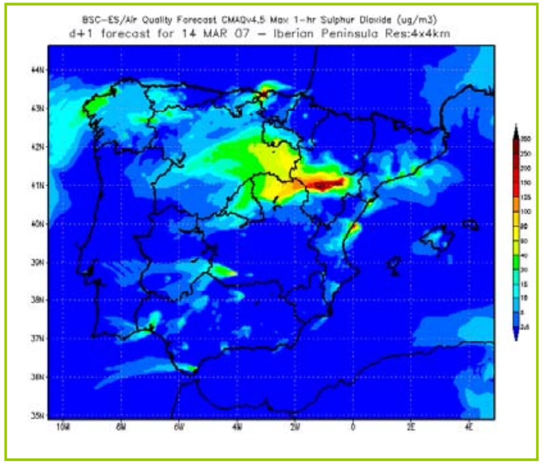


WRF-ARW
Meteorological model

HERMES
Emissions model

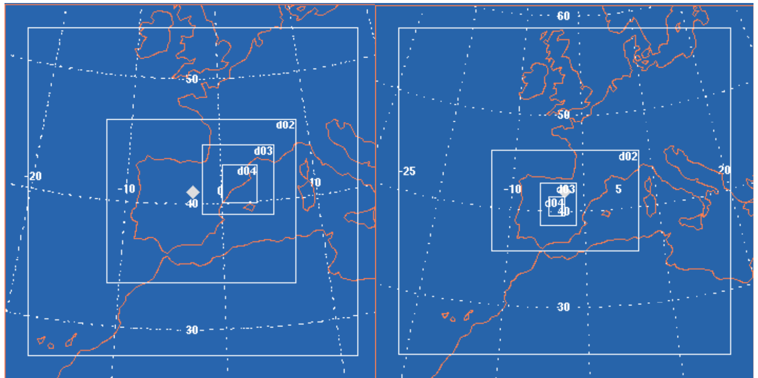


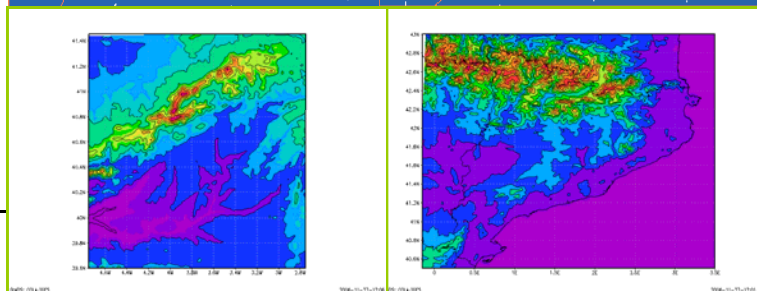
Chemistry transport model
CMAQ



Initial and boundary conditions
Nested domain in the Iberian Peninsula using EMEP data
48-h spin up

32 sigma vertical layers
4 one-way nested domains
- D1 Europe: 55x55 cells of 54km
- D2 Iberian Peninsula: 94x82 cells of 18 km
- D3 Iberian Peninsula Area: 104x103 cells of 6 km
- D4 North Eastern Domain 322x259 1 km^2 cells
- D4 Central Domain:181x214 1 km^2 cells





CMAQ model configuration

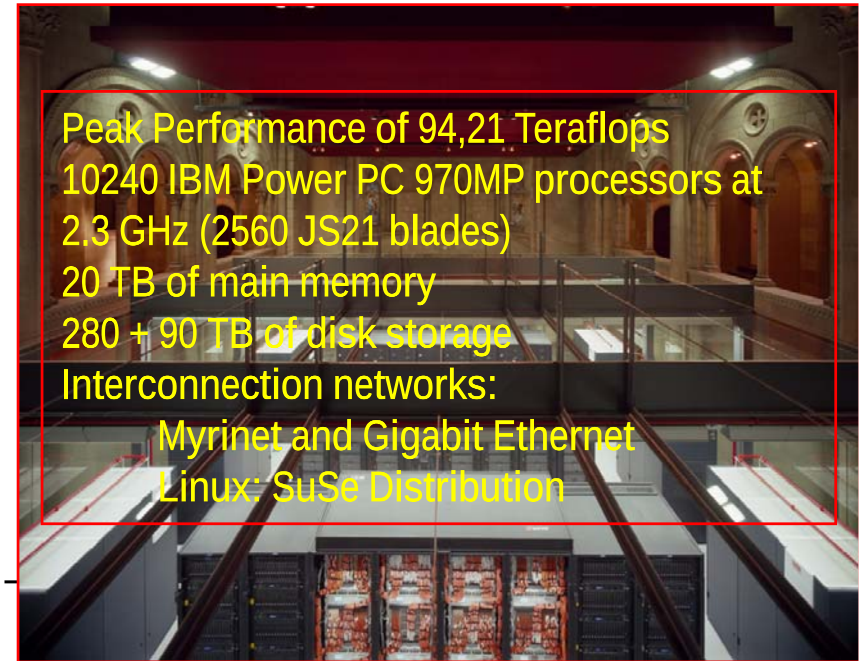
- ✓ Advection and convection scheme: Yamartino-Blackman Cubic scheme (YAM).
- ✓ Vertical and horizontal diffusion based on the Eddy diffusion scheme.
- ✓ Chemical mechanism: Carbon bond IV
- ✓ Aerosols: aero3 (3^{rd} generation modal CMAQ aerosol model)
- ✓ Aero-depv2 (2^{nd} generation CMAQ aerosol deposition velocity routine)
- ✓ Euler Backward Iterative solver optimized for the Carbon Bond-IV mechanism
- ✓ ADM-based cloud processor that uses the asymmetric convective model to compute convective mixing

Process Analysis- IPR: **integrated process rate analysis.** It provides the effects of all the physical processes and the net effect of chemistry on model predictions.

Computational resources: MareNostrum Supercomputer

Located in the Barcelona Supercomputing Center-Centro Nacional de Supercomputación (BSC-CNS, Barcelona, Spain)

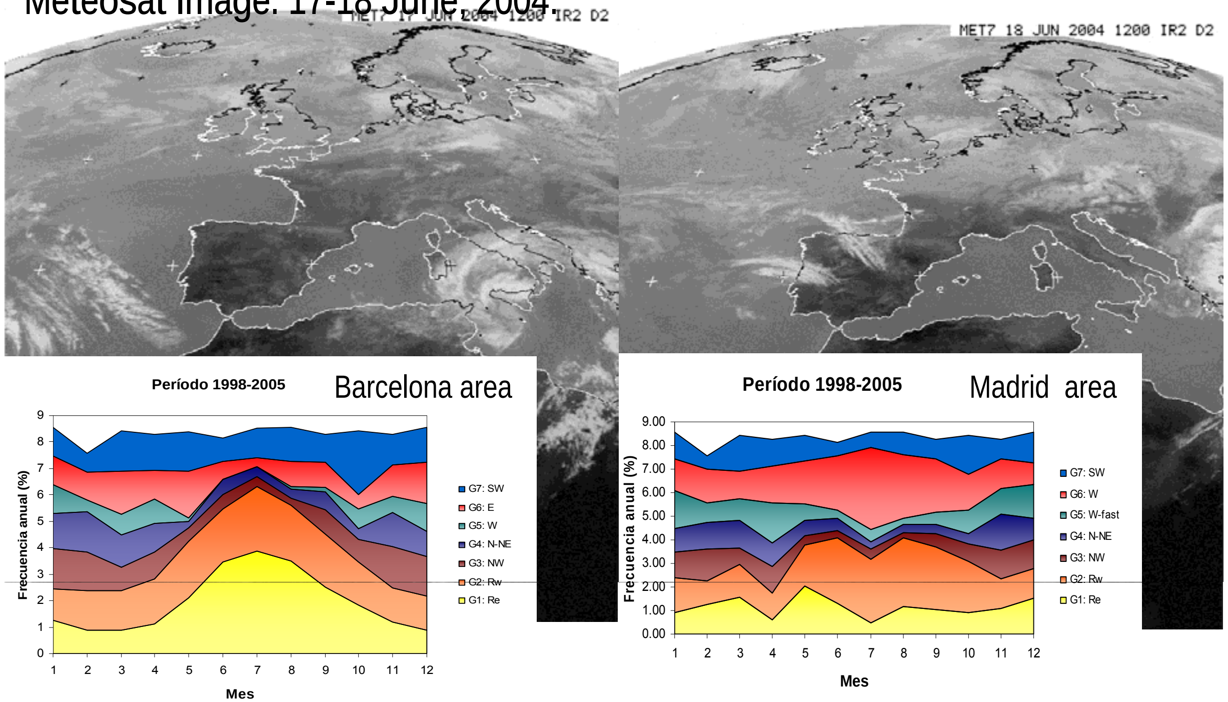
- ✓ The high spatial and temporal resolution used (1 km^2, 1hr) and the increase of the number of variables with respect to a simulation without process analysis involves large computational times.
- ✓ The simulations were feasible in time thanks to the computational power of the MareNostrum Supercomputer.



Atmospheric circulation during the episode

The weak synoptic forcing involves that the superficial wind flows are dominated by mesoscale phenomena.

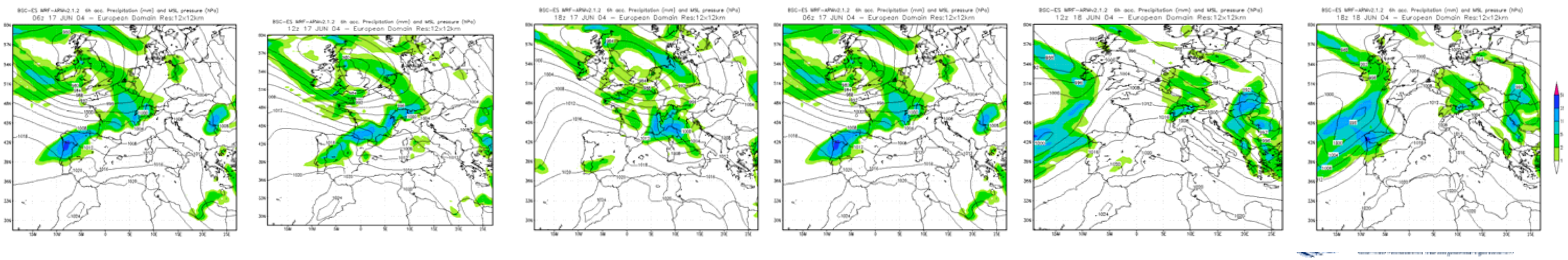
Meteosat image. 17-18 June, 2004.



- Development of the Iberian thermal low
- The atmospheric circulation in the eastern coast is dominated by land-sea breezes
- The main processes inland are the convective circulations developed by the surface heating and the formation of compensatory subsiding flows in coastal areas.

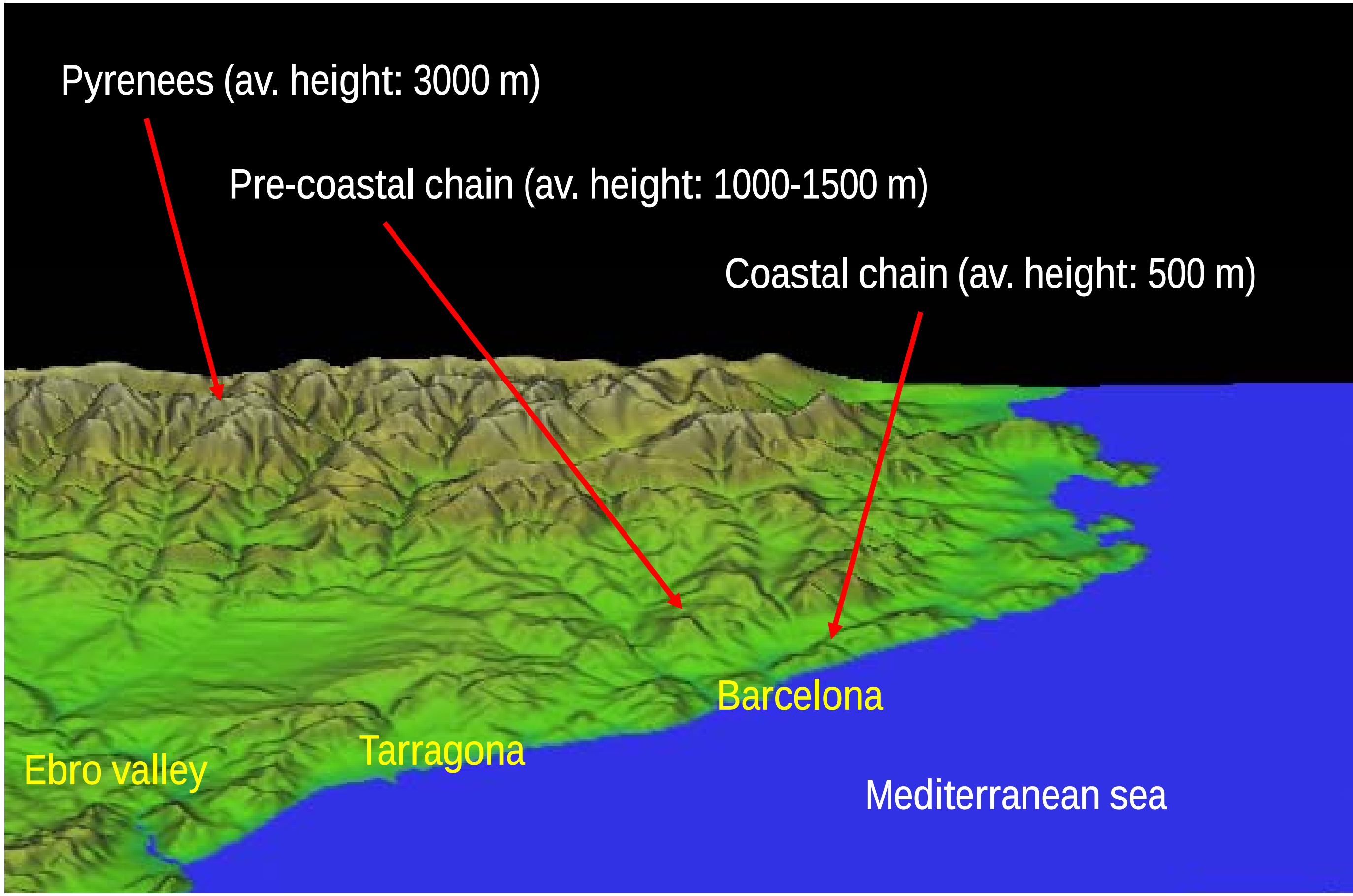
6-hr accumulated precipitation and sea level pressure for the European domain estimated by the WRF-ARW model (17-18 June, 2004).

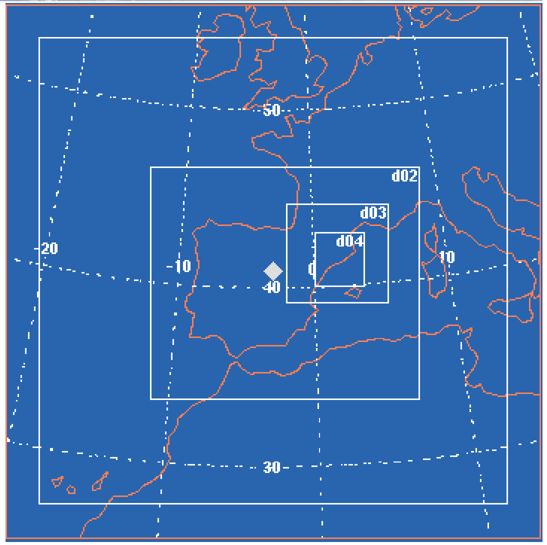
17/06/2004 06:00 | 17/06/2004 12:00 | 17/06/2004 18:00 | 18/062004 06:00 | 18/06/2004 12:00 | 18/06/2004 18:00

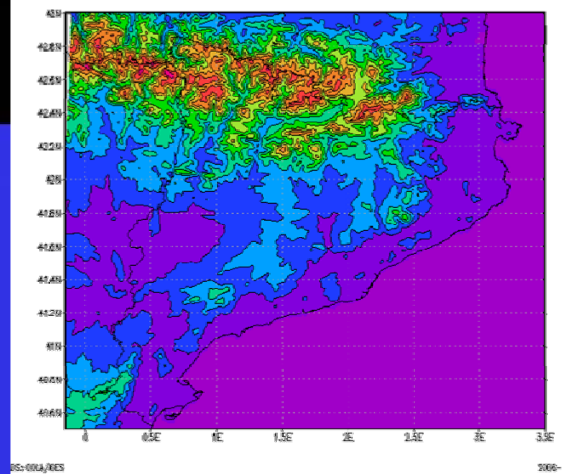


North eastern Iberian Peninsula domain. Main orographic features and anthropogenic emission sources

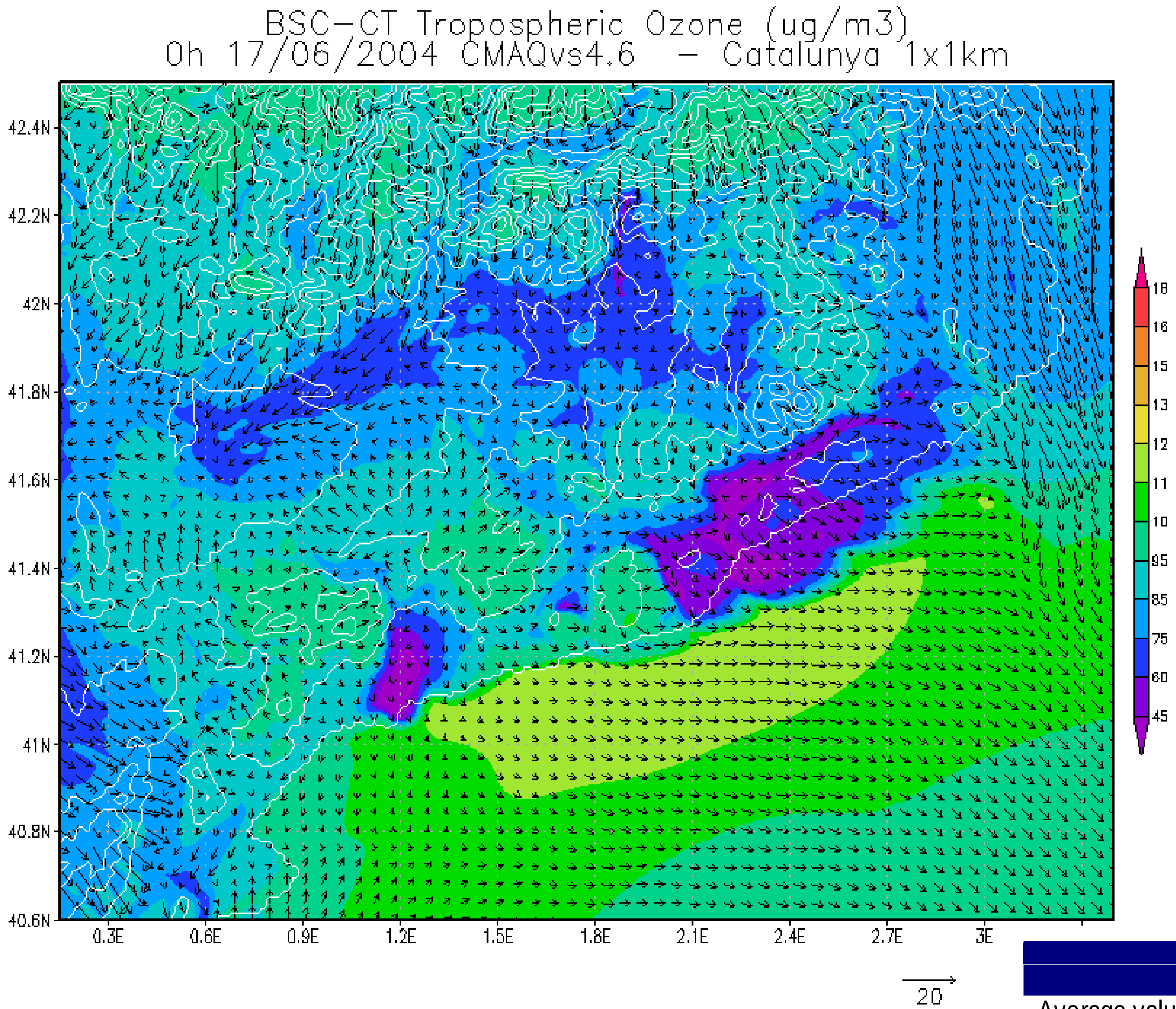
- The topography is characterized by the coastal and pre-coastal mountain chains parallel to the coast line. In the northern region the pre-Pyrenees and Pyrenees mountains have peaks higher than 3000 m. In the S-SE region the Ebro valley directly affect the winds canalization.







Surface O_3 concentration. 17-18, June, 2004.



- The sea breezes during the day and the katabatic winds and land breezes during nightime control the superficial flows.
- O_3 peaks appear from 12:00 to 16:00 UTC in the Barcelona urban plume
- It affects mainly inland areas in the north eastern and northern domain, causing exceedances of the European target (180 µg m^{-3}, 1-hr average) for human health protection.

Validation

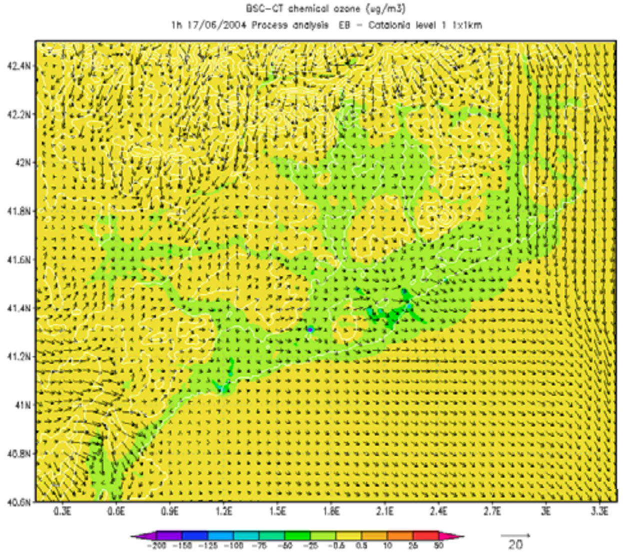
	MNBE (%)	*MNGE (%)*	*UPA (%)*
Average values (33 AQS)	-11.10%	24.73%	-16.80%

Contribution of atmospheric processes to O_3 net concentration in the North Eastern IP domain. Surface level. 17-18, June, 2004.

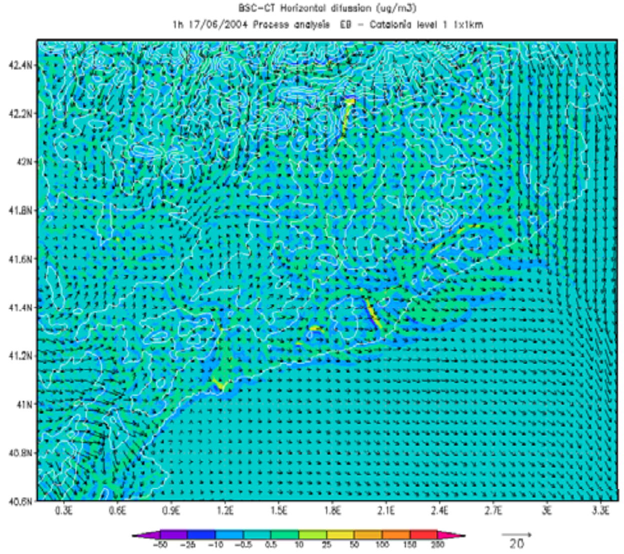
17-06-2004 0:00 UTC

- The formation of O_3 at surface level is limited to Barcelona downwind areas during the central hours of the day (from 1100UTC to 1600UTC)
- Chemical quenching of O_3 occurs at surface level at main NO_x emissions sources (roads along the coastal axis, Barcelona and Tarragona urban areas, and specific industrial areas inland)
- The O_3 concentration gradient generated involves vertical diffusion fluxes to these areas. Horizontal diffusion contributions have a lower extent.
- Wet deposition and clouds processes considered in CMAQ do not contribute appreciably to net O_3 concentration (summertime episode).

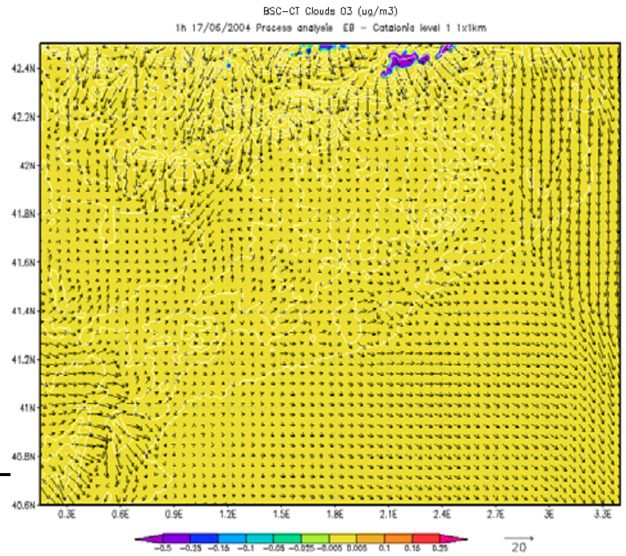
Gaseous phase chemistry (µg m^{-3})



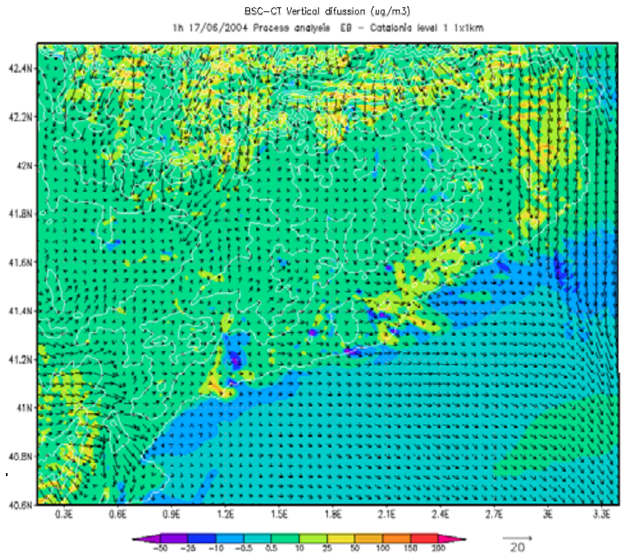
Horizontal diffusion (µg m^{-3})



Clouds processes and wet deposition (µg m^{-3})



Vertical diffusion (µg m^{-3})

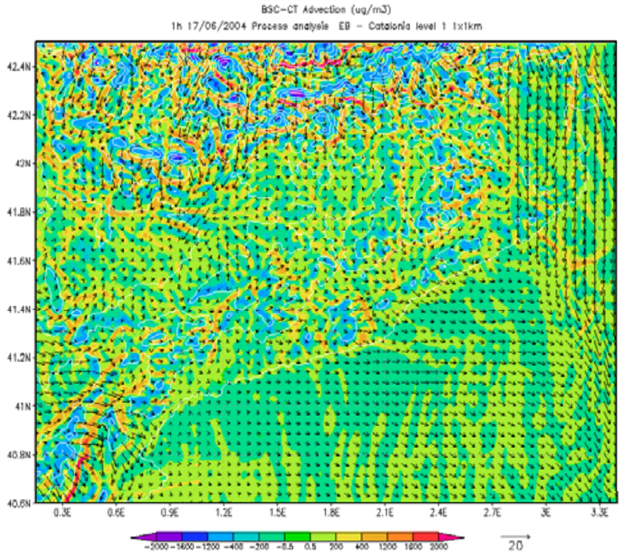


Contribution of atmospheric processes to O_3 net concentration in the North Eastern IP domain. Surface level

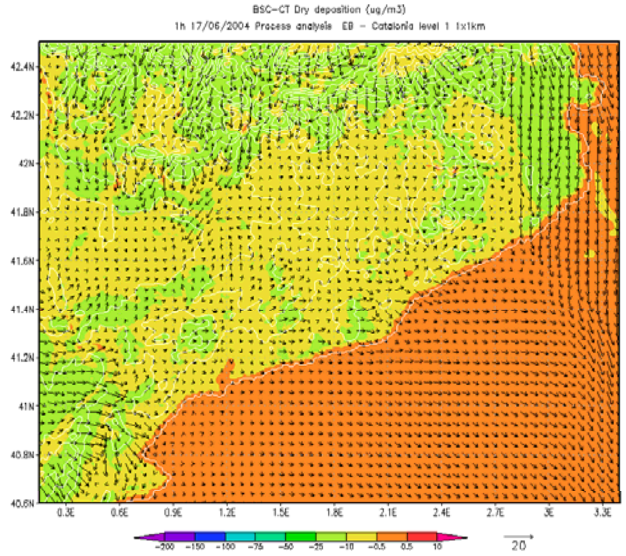
17-06-2004 0:00 UTC

- The advection and convection involve the transport of the largest amount of O_3 among the processes studied (around 10 times larger than chemical or diffusive contributions)
- A net transport balance (convection+advection contributions) permits to define the net flow of O_3 caused by wind, resulting in a term of the same order of magnitude than chemical or diffusive contributions
- The valleys and the road network channel O_3 flows caused by sea breezes during daytime and by land breezes and katabatic winds during night time.
- The dry deposition processes involve a sink of O_3 during daytime mainly in the central plateau, coastal areas and zones directly affected by the urban plume in the N-NE region

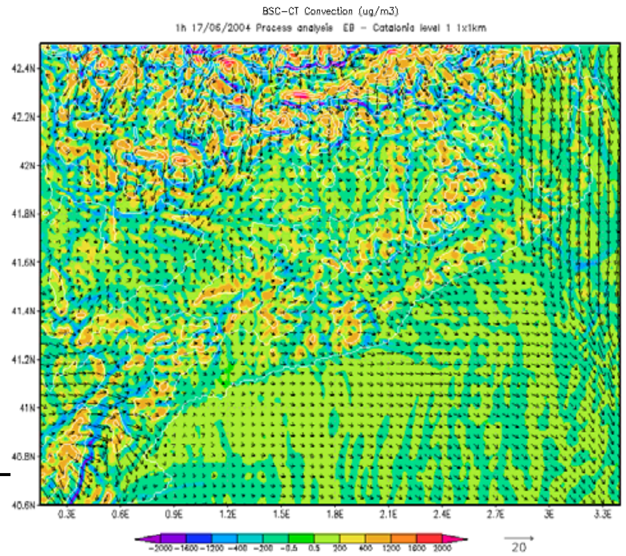
Advection (µg m^{-3})



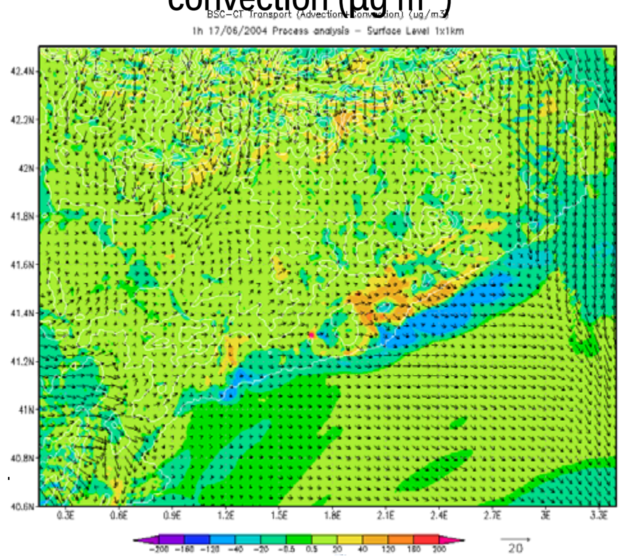
O_3 dry deposition (µg m^{-3})



Convection (µg m^{-3})



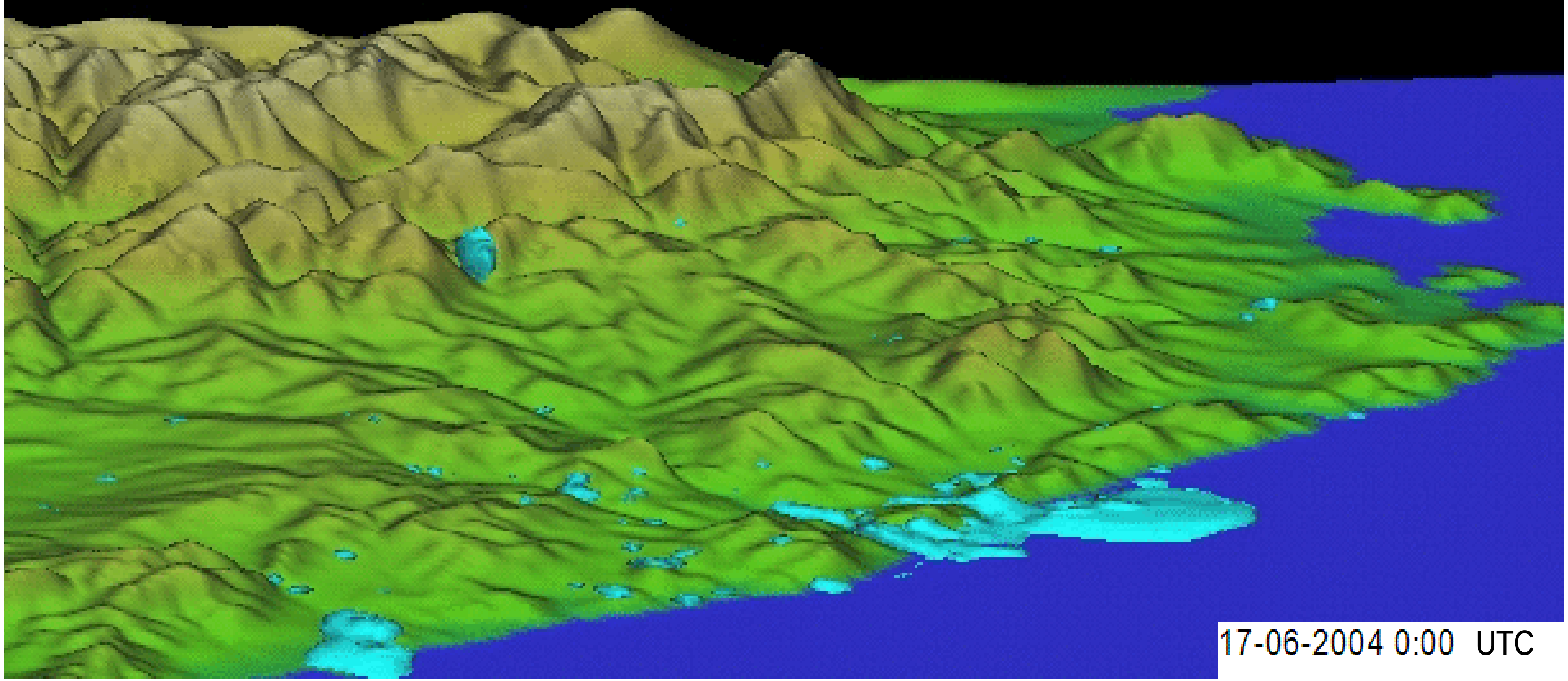
O_3 net transport: advection + convection (µg m^{-3})



Chemical ozone isosurface
17-18 June, 2004 TSTEP 1-hr

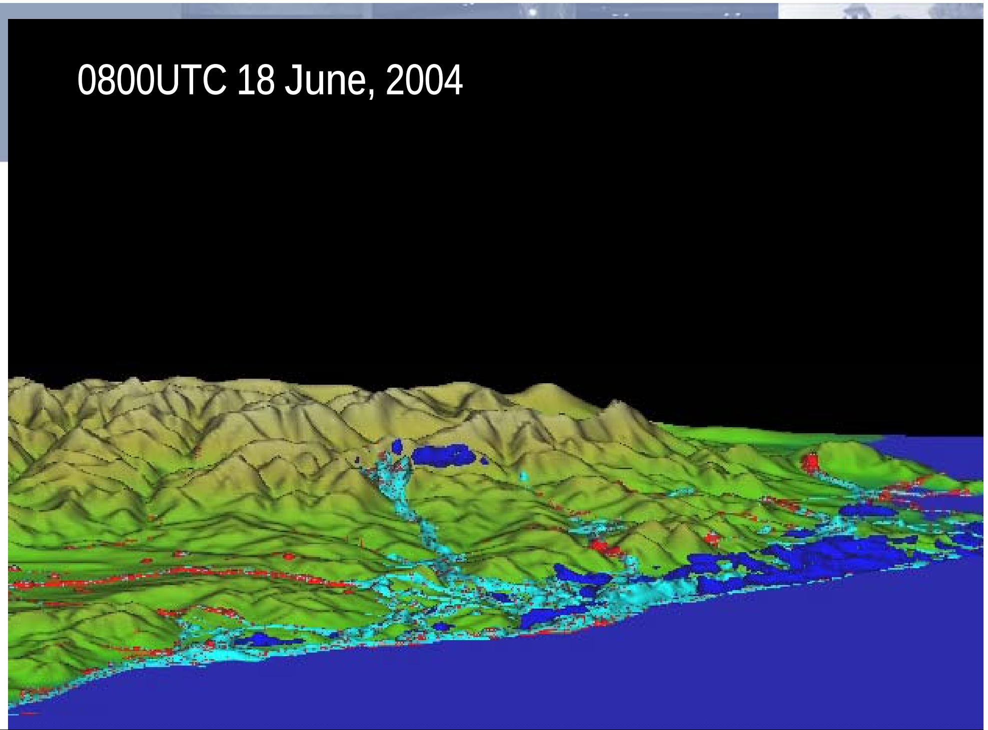
Production Red +10 $\mu g\ m^{-3}$
Destruction Blue -10 $\mu g\ m^{-3}$

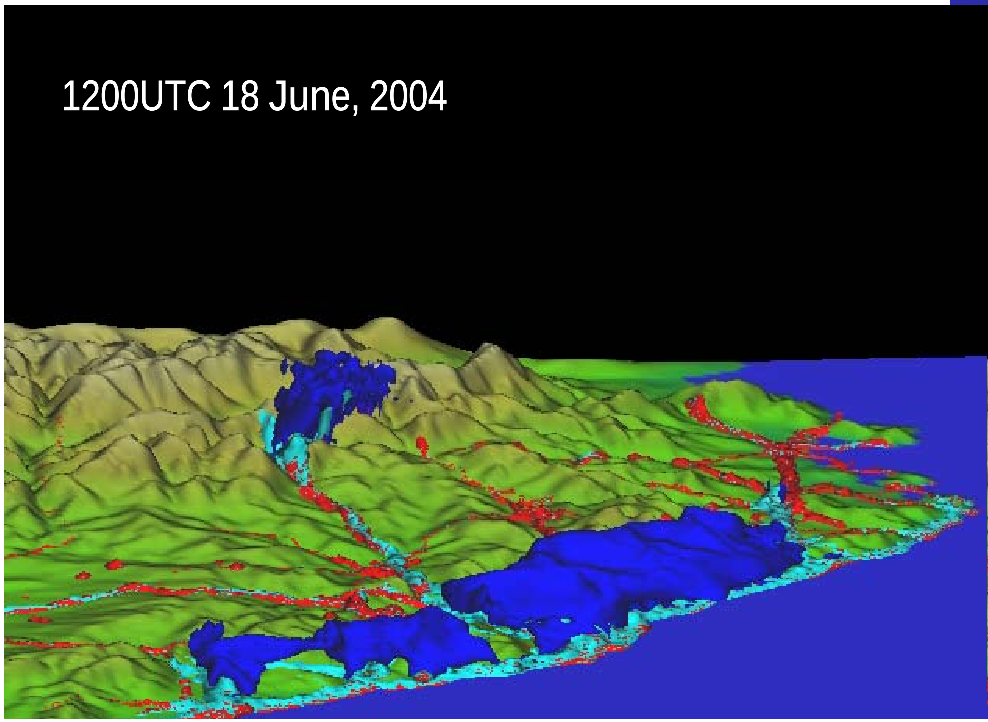
Formation of multiple layers above the mixing layer of different origin. Accumulation of O_3 over the Mediterranean sea during the night

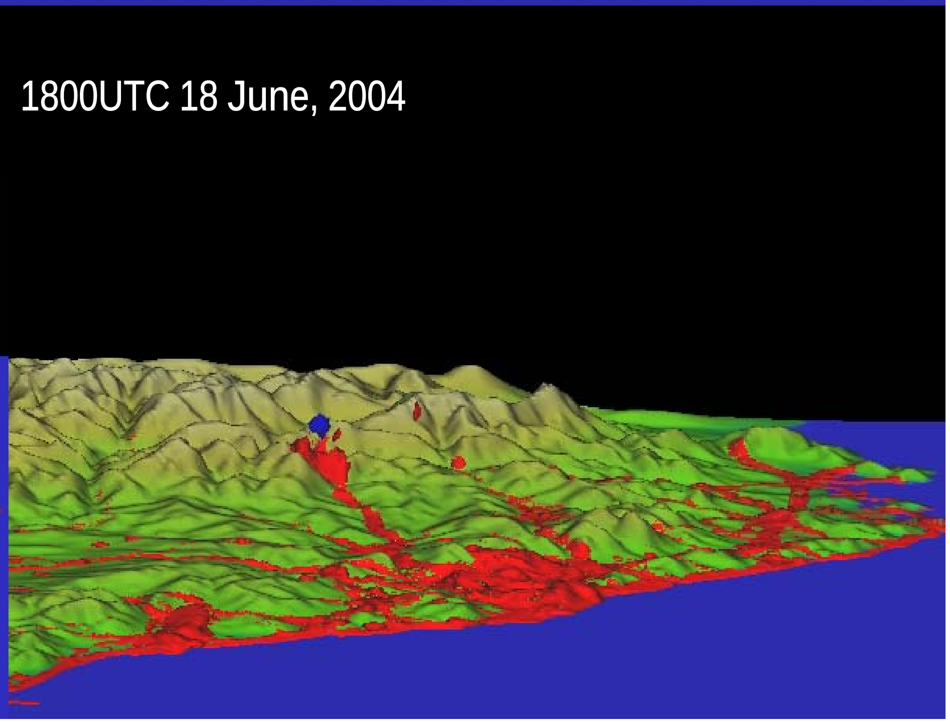


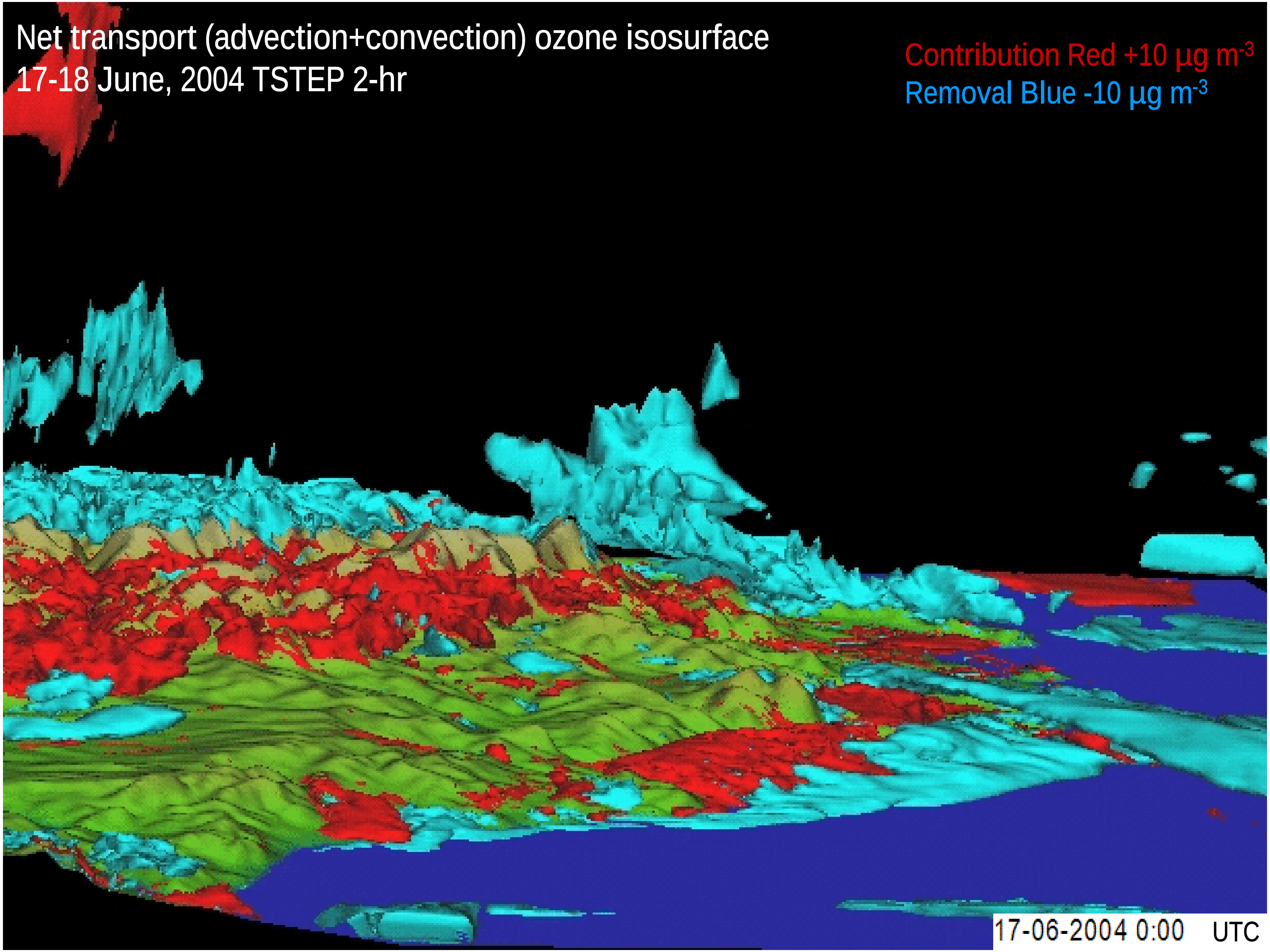
Destruction of NO Pale blue -2 µg m^{-3}
Production of NO_2 Red +2 µg m^{-3}
Destruction of NO_2 Blue -2 µg m^{-3}

The NO emitted mainly by traffic and industry reacts forming NO_2 and acts as a local sink of O_3. Over the coastal area and inland in the Cercs power generation plant plume, the NO_2 is destroyed during the day. The NO_2 destruction process leads to tropospheric O_3 formation








Net transport (advection+convection) ozone isosurface
17-18 June, 2004 TSTEP 2-hr
Contribution Red +10 µg m-3
Removal Blue -10 µg m-3
17-06-2004 0:00 UTC

Quantitative analysis: domains definition

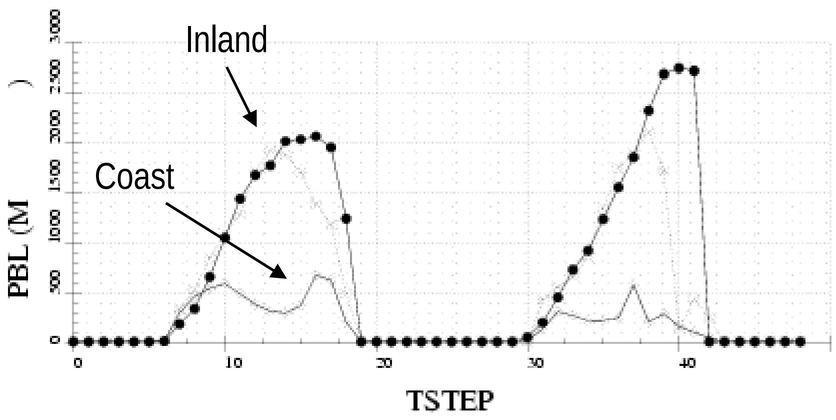
Six control volumes were defined in the north eastern IP domain in order to perform the processes outcome to O_3 net concentration.

In the horizontal plane two domains are selected based on main topographic features that affect the mesoscalar flows (coastal area and central plateau).

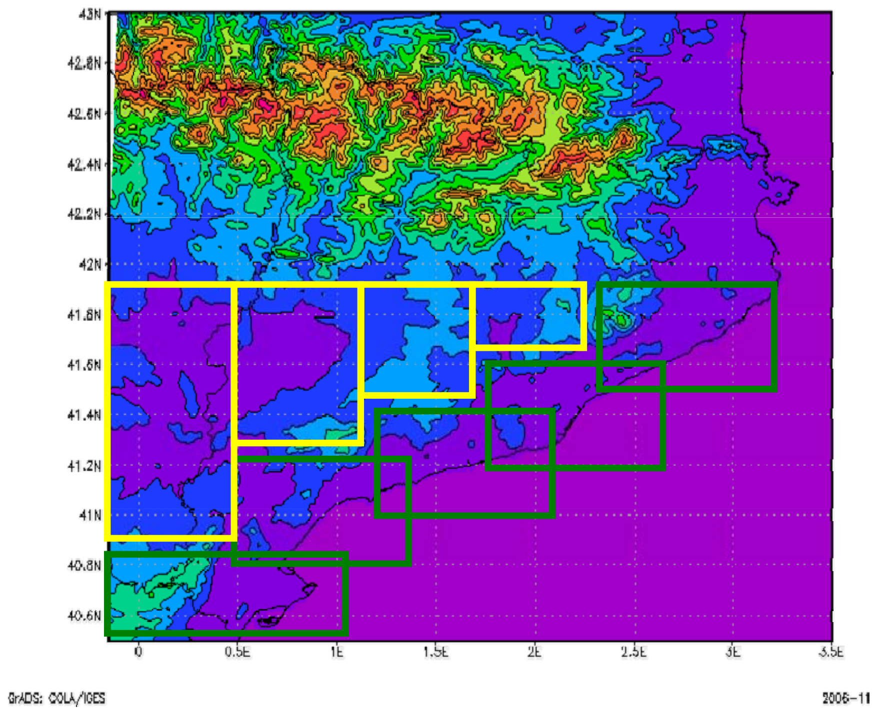
The vertical distribution pretends to understand main flows in the PBL region and the middle troposphere. The first level covers up to 500 m agl. A second level up to 1500 m agl is defined and finally the highest level top is selected at 5500 m agl.

- Horizontal domains
  - Inland domain (yellow)
  - Coastal domain (green)
- Vertical distribution
  - From 0 to 529 m agl.
  - From 529 to 1509 m agl.
  - From 1509 to 5464 m agl.

PBL height (m) in selected points of the domain



Schematic representation of the selected domains



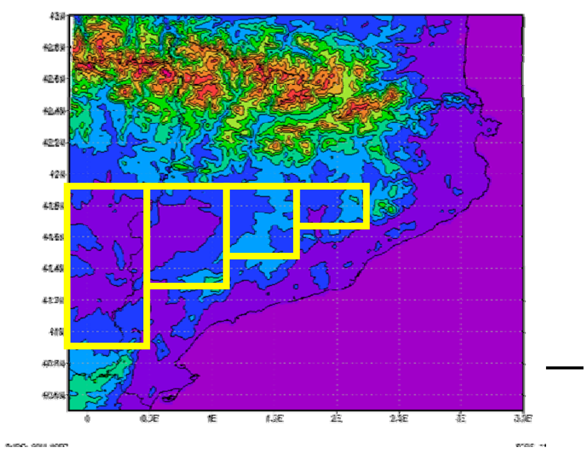
Barcelona Supercomputing Center
Centro Nacional de Supercomputación

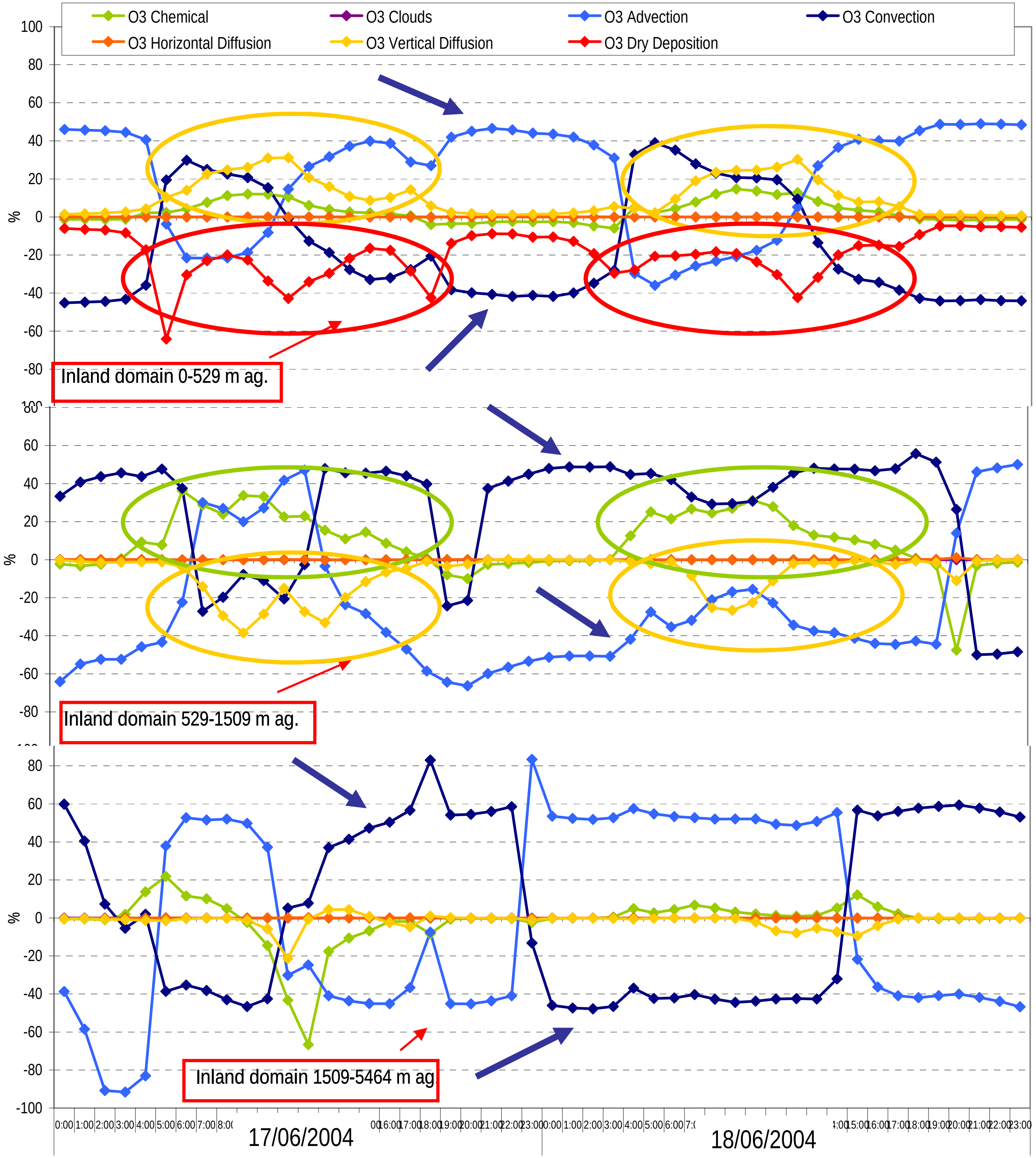
Inland domain behavior

The major source of O_3 below 500 m is the vertical diffusion (more than 20% of total contribution in the central hours of the day), while the dry deposition involves the largest sink in the domain (up to -60%).

The convection and advection processes represent a ± 60% of the total processes contribution, being the net effect an average positive contribution of 5% in this volume.

The major chemical formation (contribution larger than 30% in the central hours of the day) is produced in the 500-1500 m range.



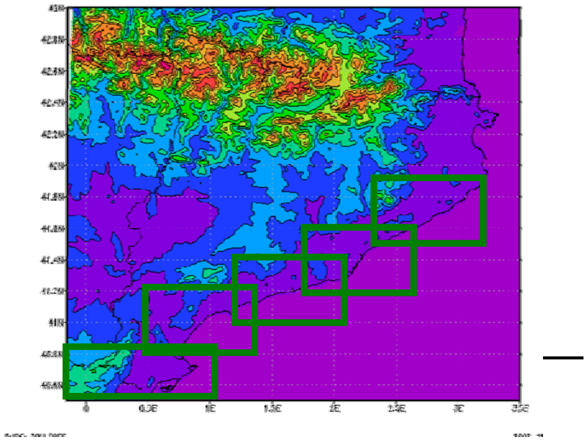


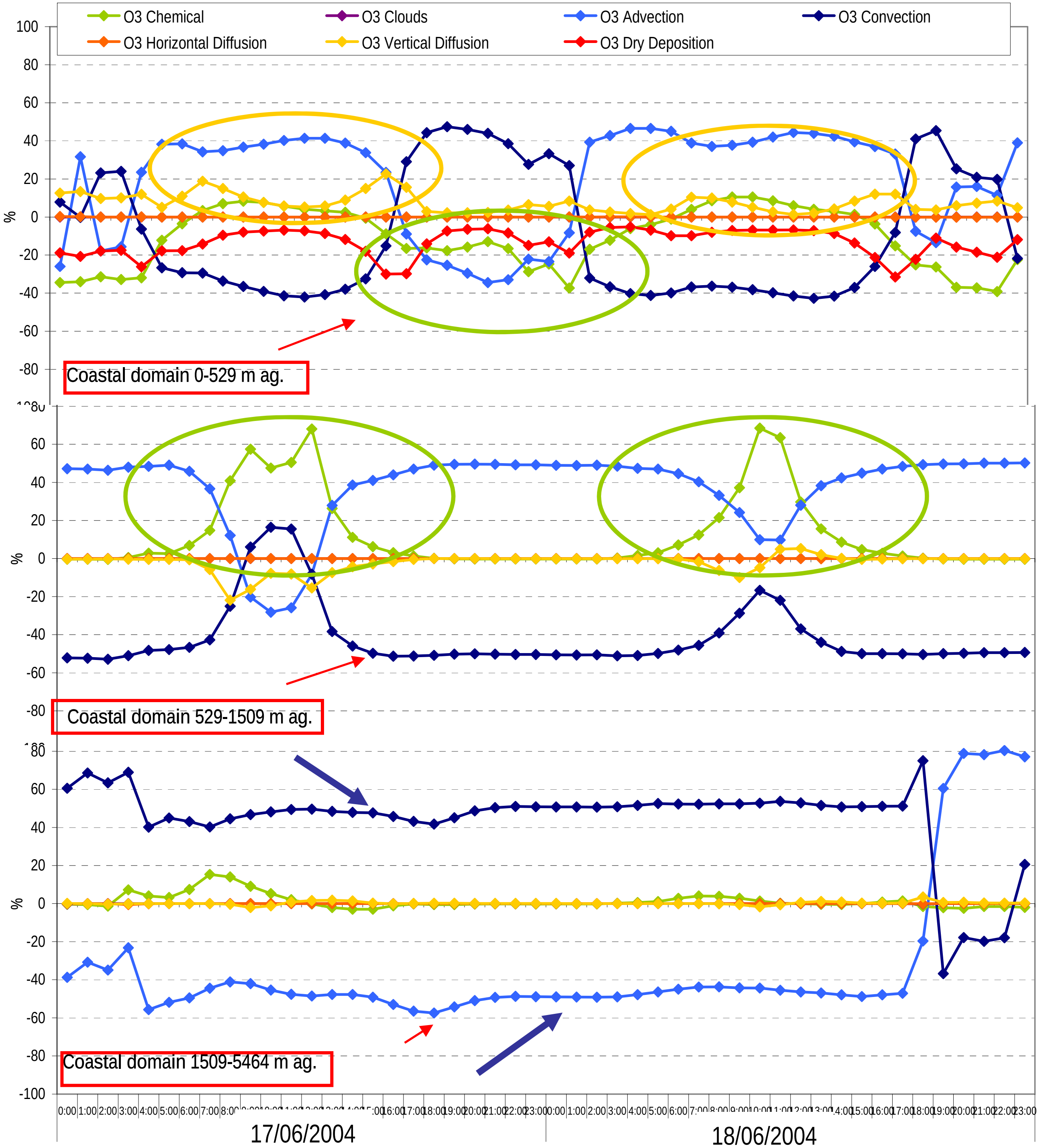
Coastal domain behavior

The large anthropogenic emission sources located in the coastal area involve the low rate of chemical formation in the first 500 m, where major contributions to net O_3 come from vertical diffusion.

The chemical formation of O_3 occurs mainly in the 500 to 1500 m agl range, where involves in the central hours of the day more than a 60% of the total contribution. Moreover the net transport in this domain is negative suggesting that not only the vertical diffusion can contribute to the lowest domain concentration but also the convection processes developed.

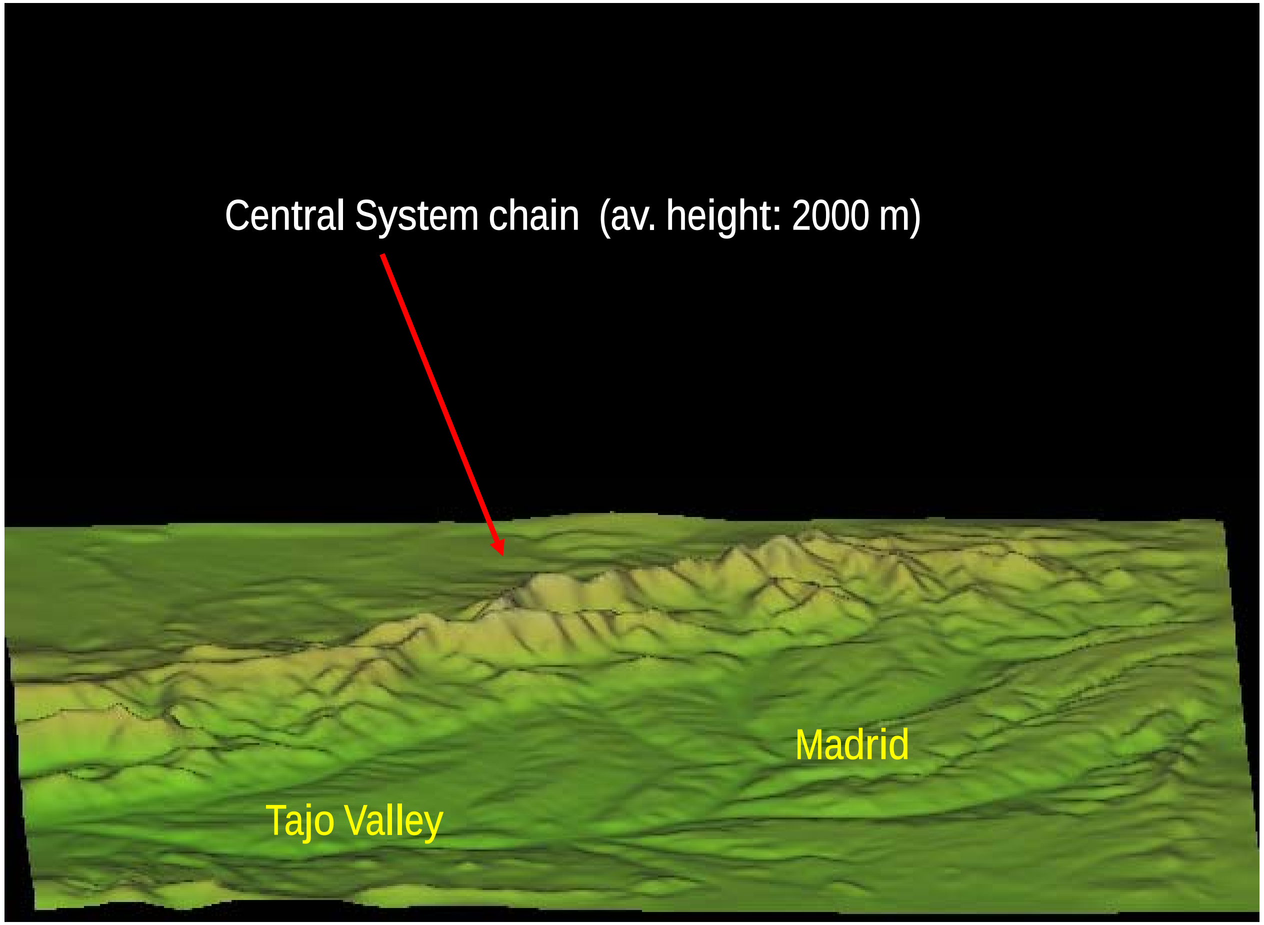
From 1500 to 5000 m transport processes dominate.

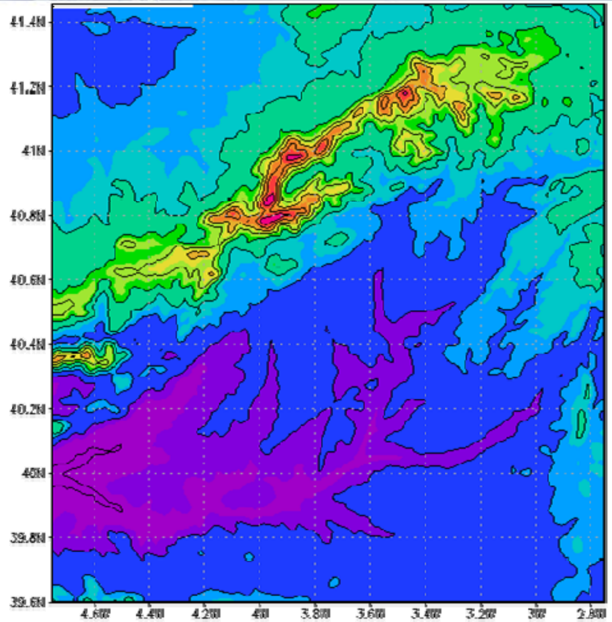


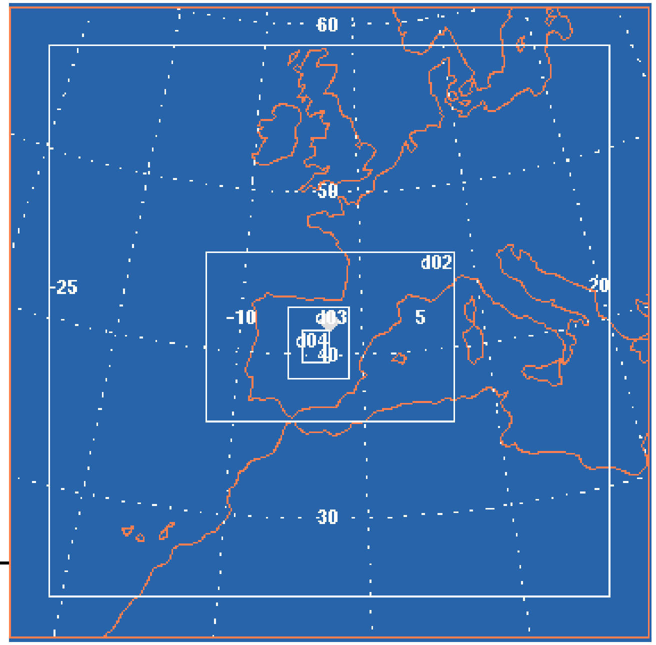


Central Iberian Peninsula domain. Main orographic features and anthropogenic emission sources

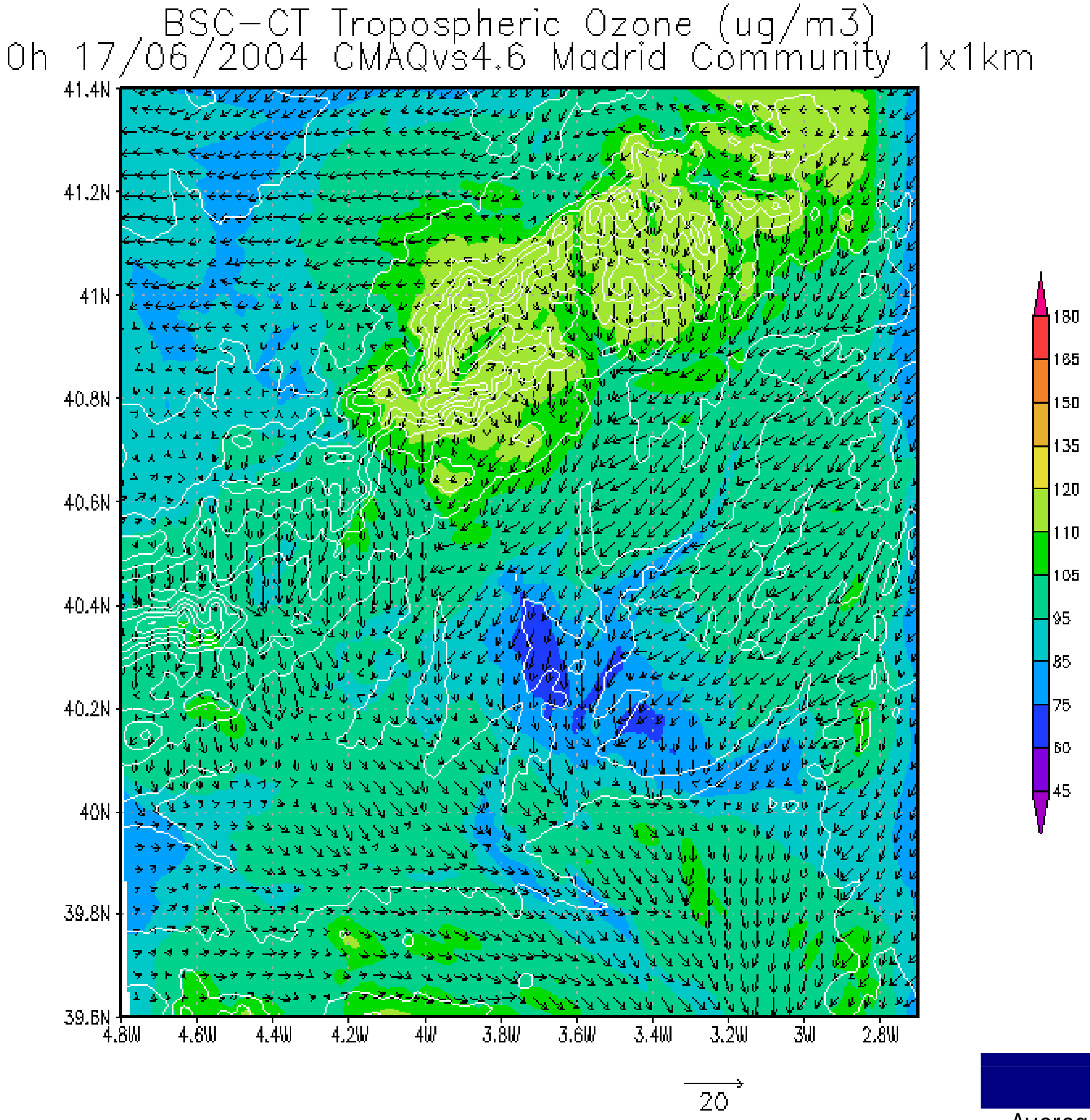
The Central System chain, with average heights of 2000 m, crosses the northern area of the domain. The southern region is characterized by the Tajo river valley. The Madrid urban area covers the center of the domain.







Surface O_3 concentration. 17-18, June, 2004.



- The superficial flows during night time are controlled by the katabatic winds in the Central System region and by the drainage along the Tajo valley.
- During the day the O_3 is mainly formed in the urban plume that is transported in the southeast direction the 1st day of the episode and to the east during the 2nd day.
- The O_3 peaks occur from 12:00 to 17:00 UTC in Madrid downwind areas.

Validation

	MNBE (%)	MNGE (%)	UPA (%)
Average values (5 AQS)	-4.29%	23.36%	13.50%

Contribution of atmospheric processes to O_3 net concentration in the Central IP domain. Surface level. 17-18, June, 2004.

17-06-2004 0:00 UTC

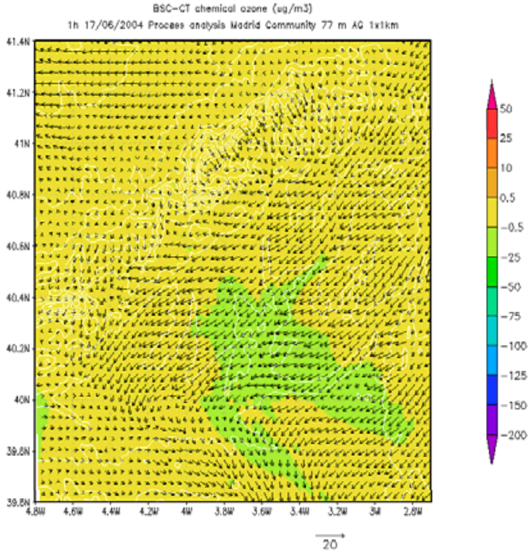
- The chemical behaviour of O_3 in the surface level is controlled by the Madrid urban area and the road network surrounding it. The O_3 destruction during the day occurs in the urban area and road network, as main NO_x emitters in the region. Nevertheless formation of O_3 in the surface level is also detected in the surrounding areas.

- Due to the O_3 concentration gradient caused by the chemical destruction a vertical diffusion flow contributes to O_3 net concentration in this surface areas.

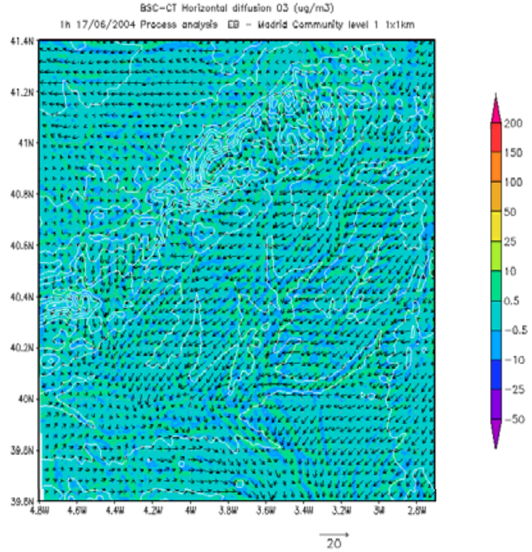
- The horizontal diffusion flows contribution to net O_3 surface is lower in an order of magnitude (10 times lower).

- The clouds processes and wet deposition are negligible compared to the rest of processes.

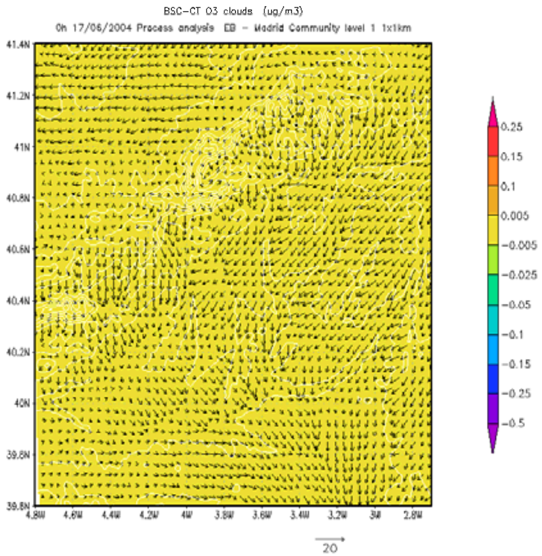
Gaseous phase chemistry (µg m^{-3})



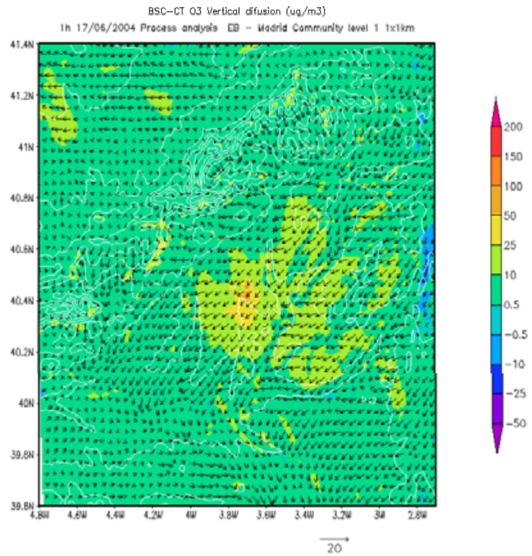
Horizontal diffusion (µg m^{-3})



Clouds processes and wet deposition (µg m^{-3})



Vertical diffusion (µg m^{-3})



Contribution of atmospheric processes to O_3 net concentration in the Central IP domain. Surface level. 17-18, June, 2004.

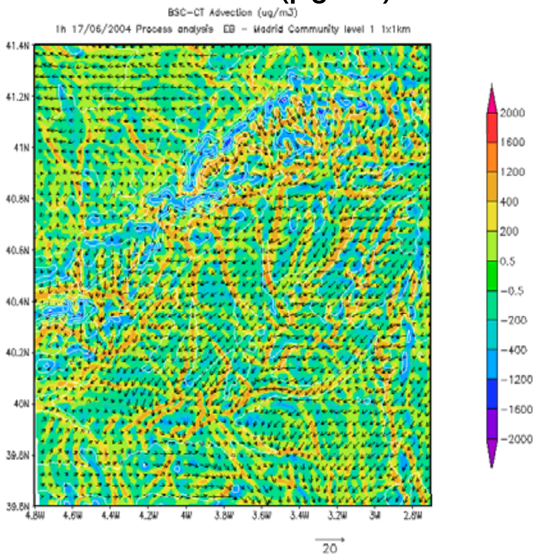
17-06-2004 0:00 UTC

- Advection and convection processes transport the largest masses of O_3 in the region. Positive and negative contributions are distributed following the topography of the region, both in the Central System and in the southern flat area forced by river valley channels.

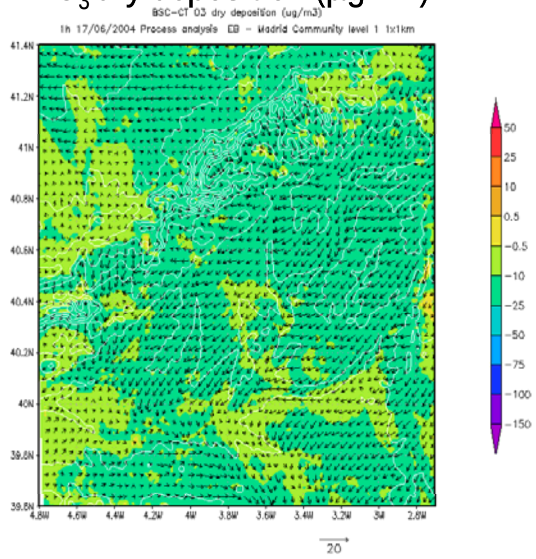
- The net effect of transport involves the O_3 masses moving from the urban airshed and surrounding areas where they are mainly formed to the southern and eastern region during the central hours of the studied days.

- The dry deposition involves an important O_3 sink, especially during daytime and in the areas where largest concentrations are achieved (downwind areas from Madrid city)

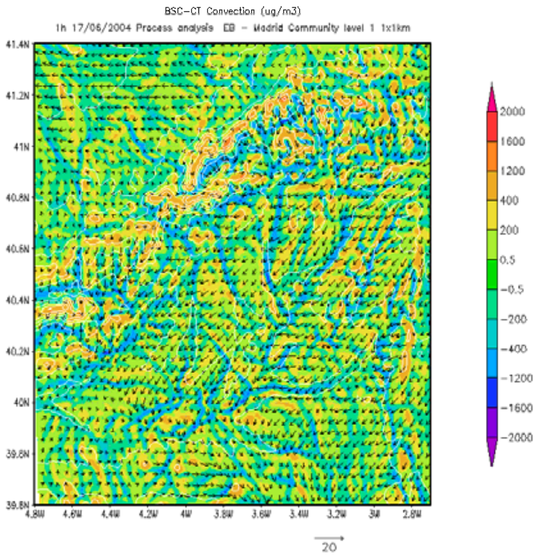
Advection (µg m^{-3})



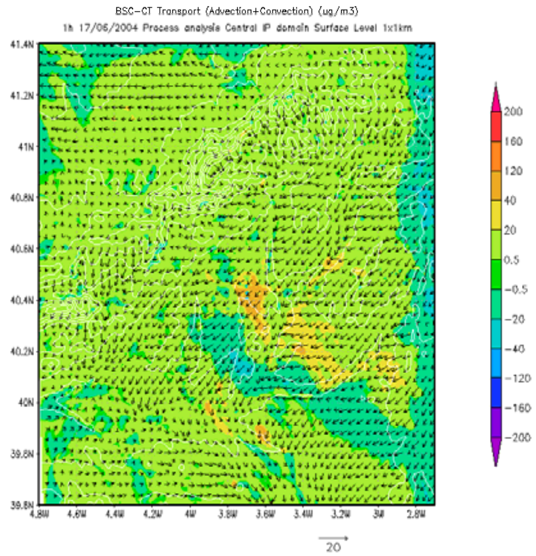
O_3 dry deposition (µg m^{-3})



Convection (µg m^{-3})



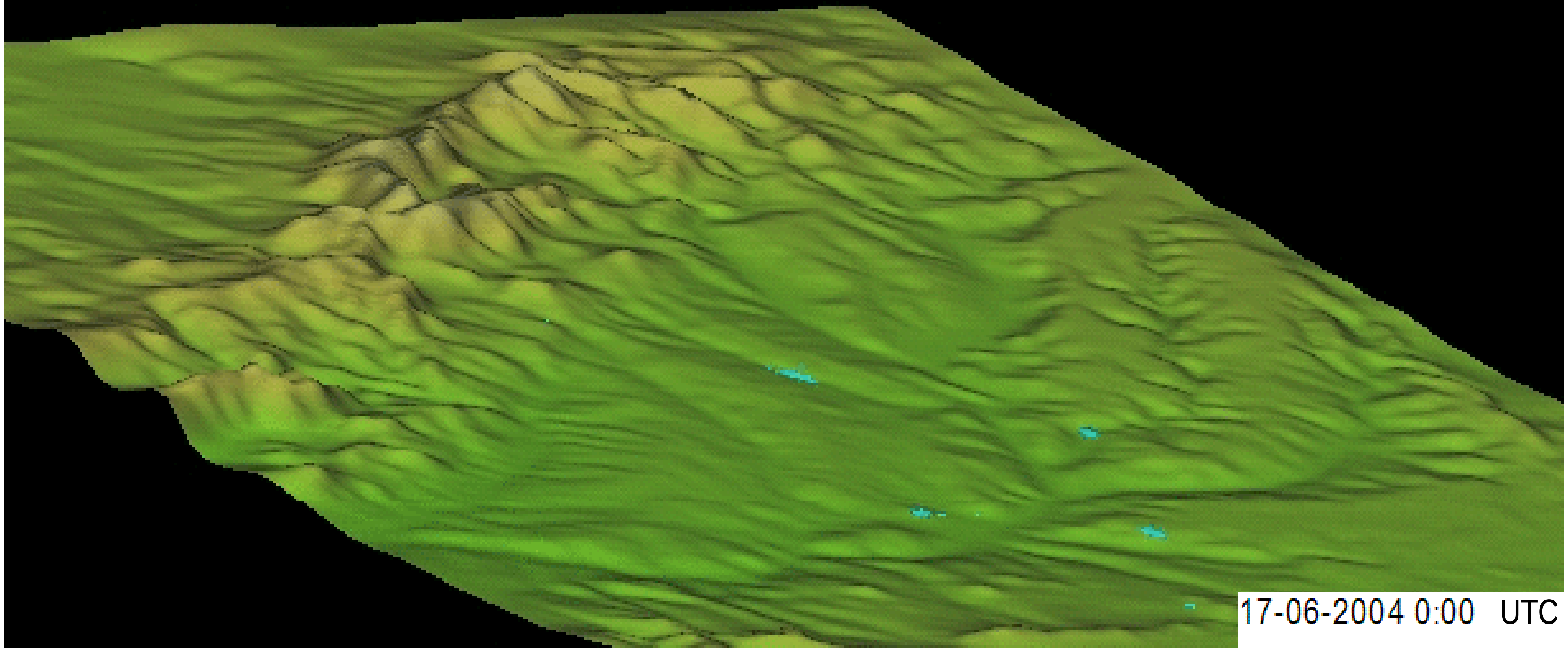
O_3 net transport: advection + convection (µg m^{-3})



Chemical ozone isosurface
17-18 June, 2004 TSTEP 1-hr

Production Red +10 µg m^{-3}
Destruction Blue -10 µg m^{-3}

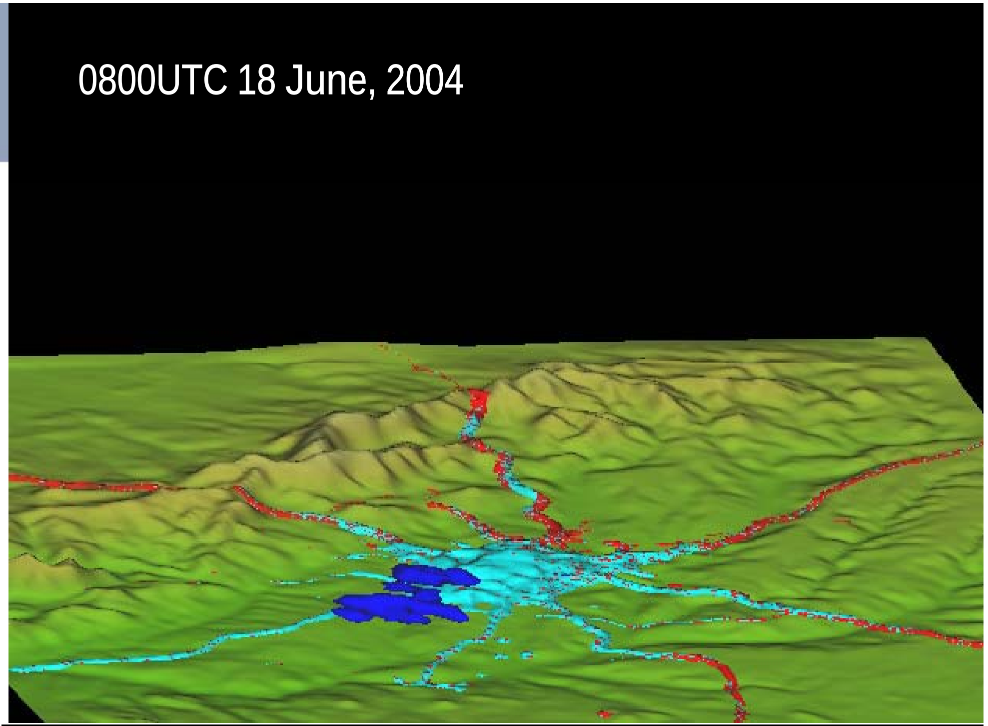
Thermal phenomena involve the urban plume elevation during the morning.
The loss of strength of the convective cell occurs at last hours of the day downwind.

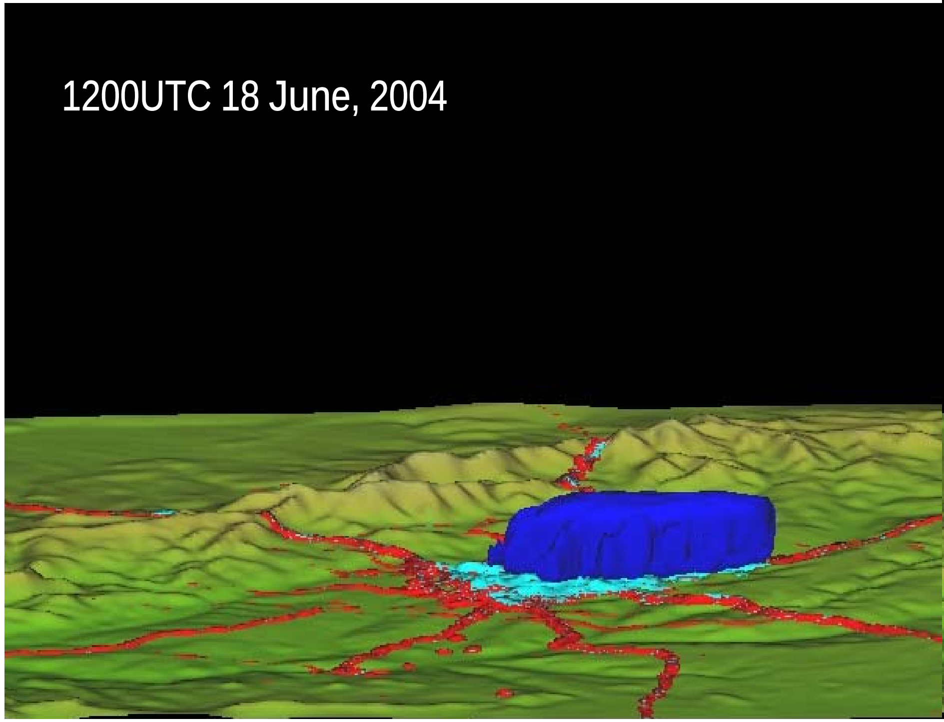


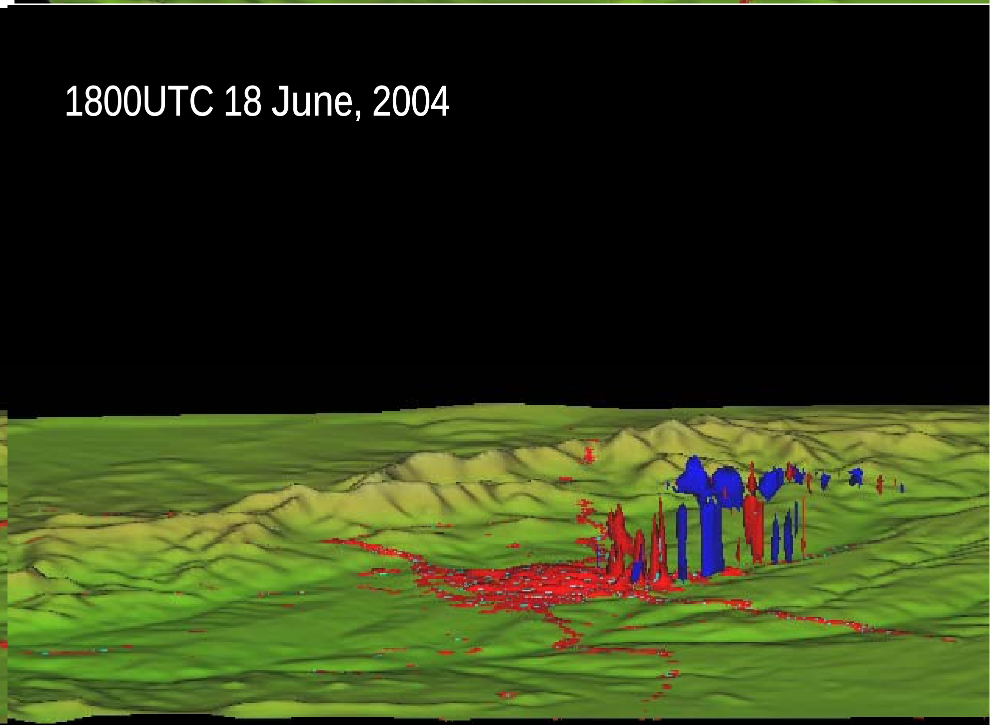
Chemical NO_x isosurfaces

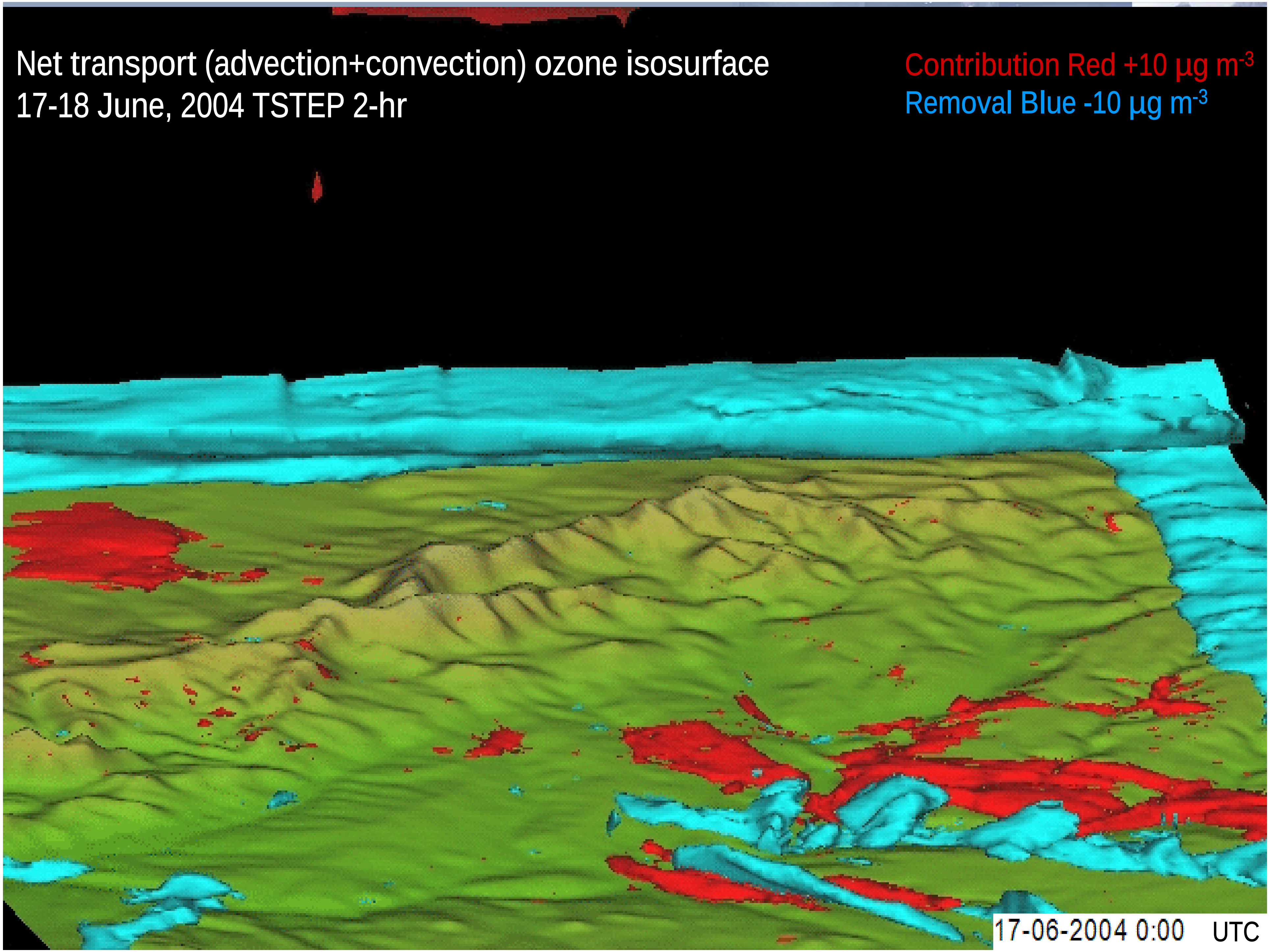
Destruction of NO Pale blue -2 μg m^{-3}
Production of NO_2 Red +2 μg m^{-3}
Destruction of NO_2 Blue -2 μg m^{-3}

The NO emitted mainly by traffic reacts forming NO_2. The chemical destruction of this compound occurs in the urban plume producing O_3.








Net transport (advection+convection) ozone isosurface
17-18 June, 2004 TSTEP 2-hr
Contribution Red +10 µg m-3
Removal Blue -10 µg m-3
17-06-2004 0:00 UTC

Quantitative analysis: domains definition

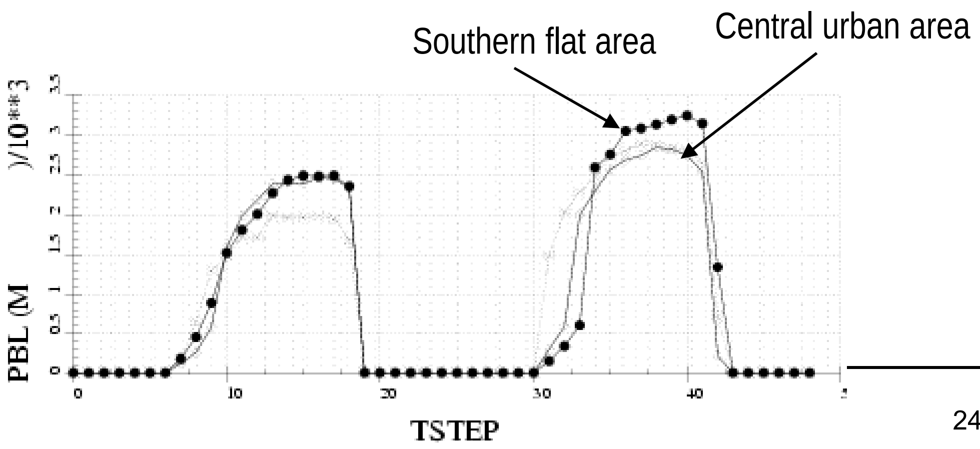
Six control volumes were defined over the central IP domain in order to better describe the processes contribution to O_3 concentrations, both in the horizontal and vertical dimension.

In the horizontal plane two regions were defined intending to describe the different behaviour caused by major emission sources: the southern flat region over the Tajo valley and the central area in which the Madrid conurbation is located.

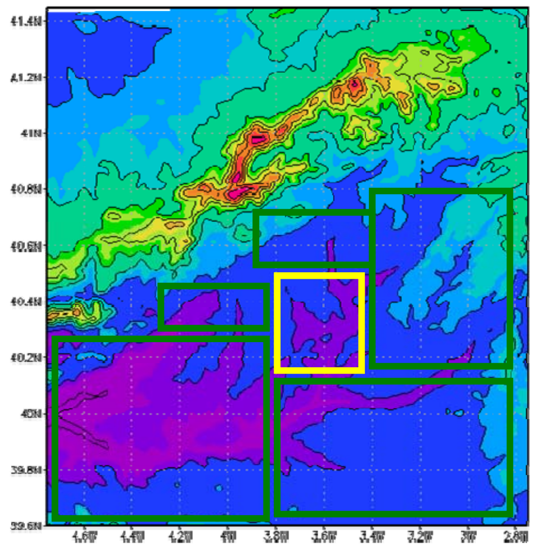
In the vertical component three ranges are defined in order to detect different behaviours under the PBL. The maximum PBL height is achieved at 16:00 UTC of the 18/06/2004 and reaches up to 3500 m agl. depending on the selected area of the domain.

- Horizontal domains
  - Flat domain (green)
  - Central (*urban*) domain (yellow)

- Vertical distribution
  - From 0 to 529 m agl.
  - From 529 to 1509 m agl.
  - From 1509 to 5464 m agl.



Schematic representation of the selected domains

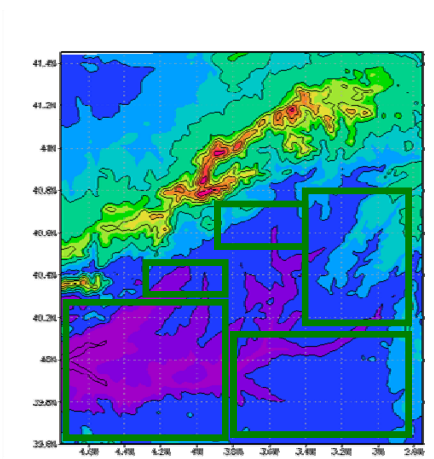


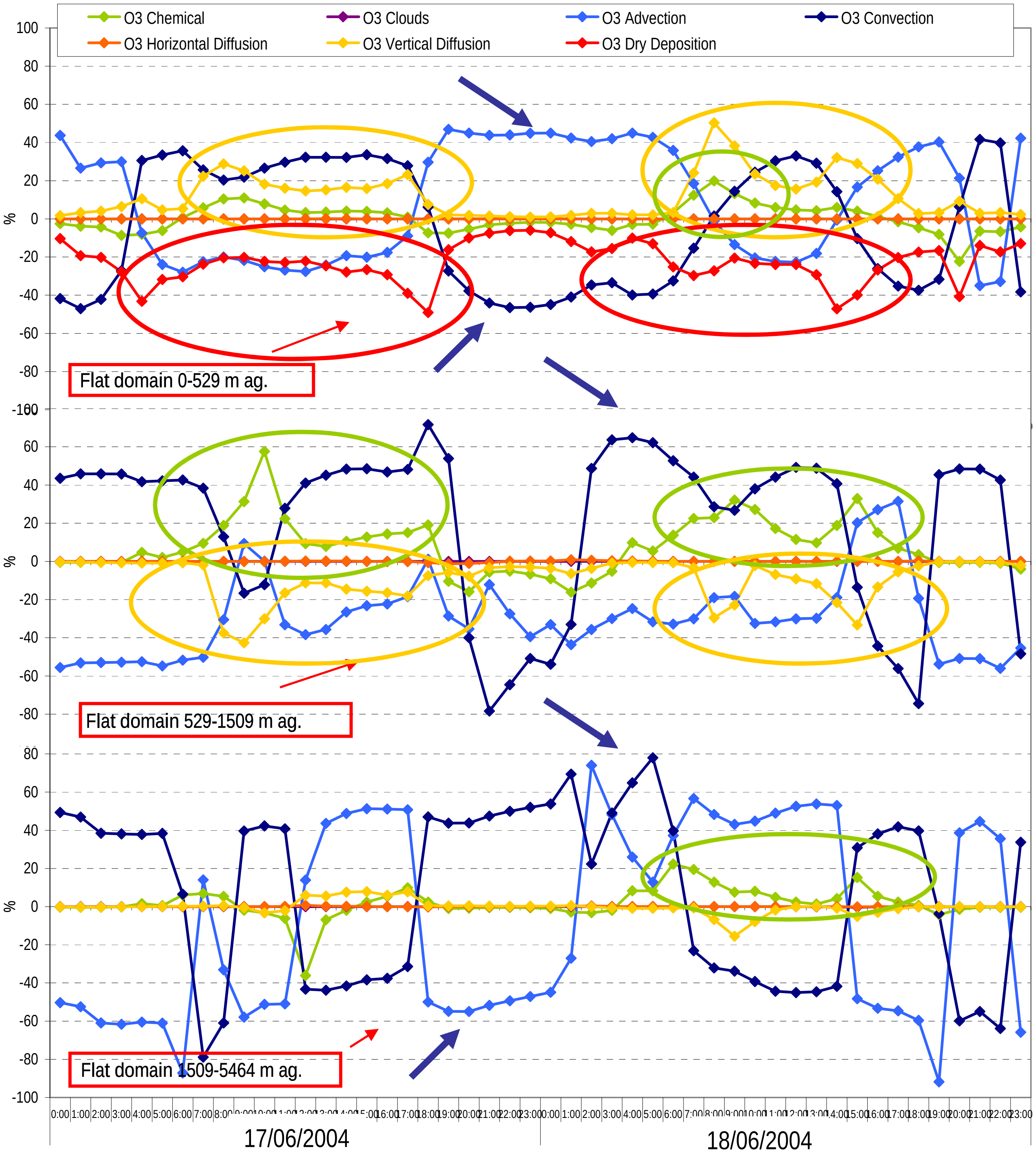
Flat domain behavior

The O_3 net concentration at surface level is caused mainly by vertical diffusion (20-40% during the day) and chemical production (up to 20% of total contributions).

The main sink of O_3 is the dry deposition, accounting for -40% of total contributions.

Advection and convection move the largest masses of O_3 in the region, their net effect involves a 6% contribution on average in the lower level (up to 500 m agl.) and a -8.8% in the 500-1500 m layers.





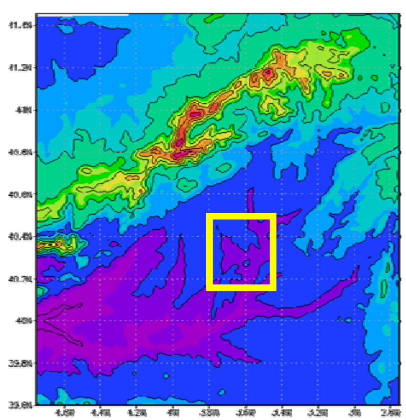
Central (urban) domain behavior

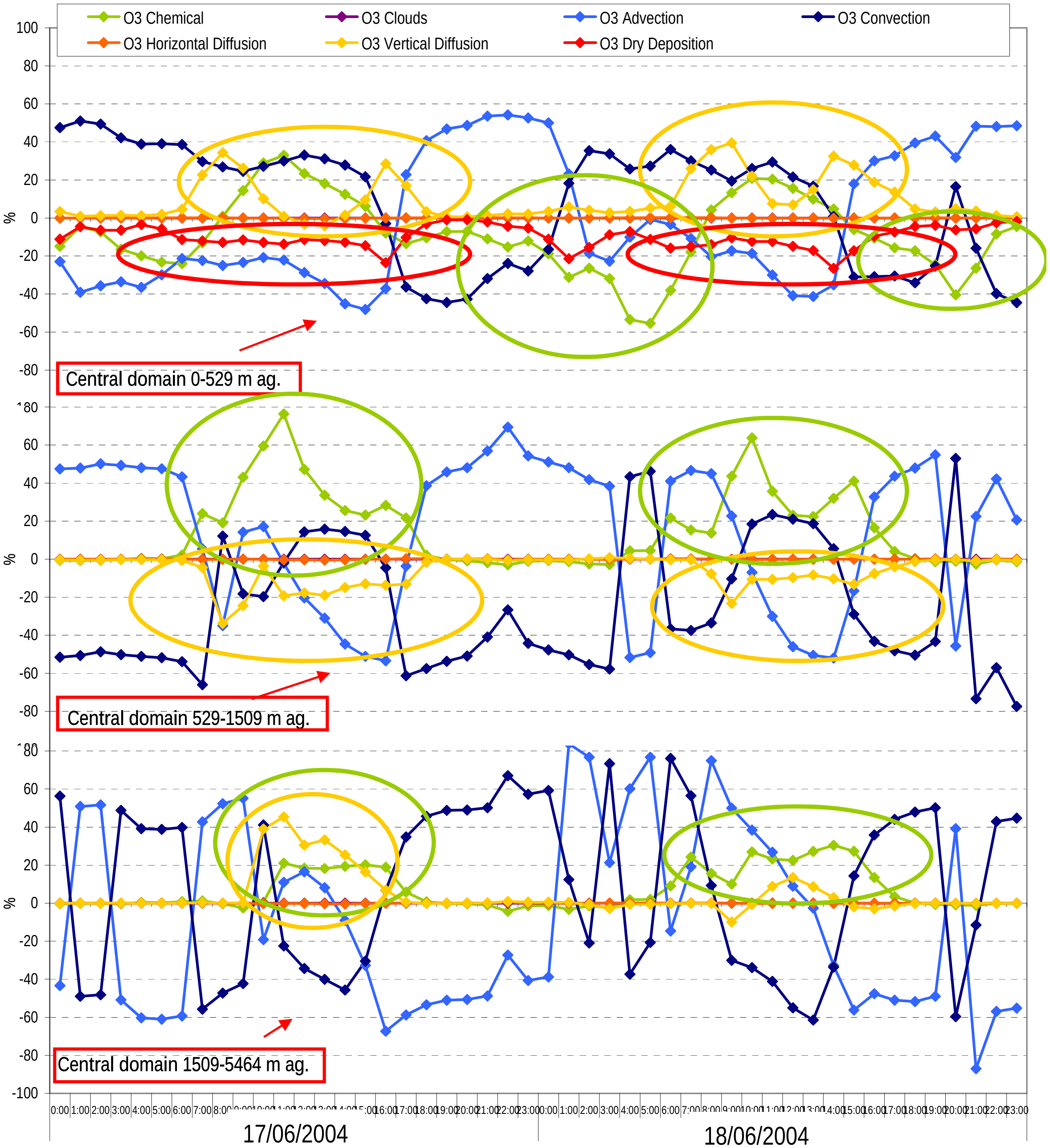
Chemical O_3 formation accounts for a 20-30% of total contributions to net O_3 in the 0-500 m agl. levels, during the central hours of the day. The largest contributor to O_3 in this volume is the vertical diffusion.

Transport by advection and convection moves the largest mass of O_3, being the net effect a 7% contribution on average.

The importance of this processes is larger in the 500-1500 m agl. level, where the net transport involves a -11% of total contributions on average. The chemical formation occurs mainly in these levels, accounting up to an 80% of net O_3 during the central hours of the 17/06.

Chemistry and diffusion processes contribute to net O_3 concentrations in the 1500-5500 m agl. zone just in the central hours of the selected days (+20-40%).





Conclusions

- The process analysis tool implemented in the CMAQ model was applied during a summertime photochemical pollution episode:17-18 June, 2004 to the central and north eastern Iberian Peninsula.

- During the episode, the mesoscalar phenomena control the flows in both regions, but the presence of the coastal line and the very complex topography of the north eastern domain determines a more complex behaviour. This is captured by the model, that provides a deep description of i.e. the breeze recirculation cells or the O_3 accumulation layers formed over the Mediterranean sea during the last hours of the day.

- The O_3 photochemical formation occurs mainly in downwind areas from the NO_x emission sources. The maximum contribution is detected in levels over the surface, where these sources involve the main sink of O_3 together with the dry deposition.

- In the central Iberian Peninsula domain the formation occurs from surface up to the 1500-5500 m agl. levels, due to the convective cell development, which is not observed for the north eastern Iberian Peninsula, where the formation is maximum at 500-1500 m agl. levels, due to the local recirculations and layering of pollutants.

- Moreover the photochemical regime involves the O_3 destruction at surface level in Barcelona urban area, the results suggest a lower NOx/VOCs ratio in Madrid where photochemical production is observed during the central hours of the day.

Thanks for your attention

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