

Transboundary pollution and local emission impact in tropospheric ozone accumulation processes: control strategy modelling assessment

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Abstract—Ozone production and accumulation in troposphere are nonlinear processes, forced by precursor emissions, either from anthropogenic or natural sources, and solar radiation. Furthermore, photochemical pollution can be locally produced or can be due to transboundary processes. Because of the problem complexity, mathematical model represents a useful tool for regional Authorities to implement efficient air quality control strategies. In this paper the factor separation approach has been formalized and applied to assess the relative and absolute boundary condition and local emission impact in tropospheric ozone accumulation processes over Northern Italy. The proposed technique processes the simulations of a 3D photochemical modelling system.

I. INTRODUCTION

Over the last decades the pollutants characterizing the atmosphere chemical composition have been changing substantially and in the recent years one of the main concerns has been photochemical pollution. Ozone is considered one of the most significant photochemical pollutants with respect to the potential impact to human health and natural ecosystems, both in terms of severe episodes and as long-term exposures. The formation and accumulation of ground level ozone concentrations are nonlinear processes, (a) depending on a large number of reactions taking place in the atmosphere, (b) favored by meteorological stagnating conditions, as elevated solar radiation and weak winds, and (c) strongly forced by primary emissions of its precursors (nitrogen oxides and volatile organic compounds). Thus, the definition of efficient ozone control strategies is a complex task, mainly ruled by the chemical regimes that characterize the domain under study [1]: (1) the NO_x-sensitive regime, in which ozone increases with increasing NO_x emissions and shows relatively little change in response to increased VOC, and the VOC-sensitive (or NO_x-saturated) regime, in which ozone increases with increasing VOC emissions and decreases with increasing NO_x emissions.

Several studies in literature are addressed to the modelling assessment of the benefits of NO_x versus VOC control (e.g. [2]), but only few investigate the boundary conditions effects on control strategy (e.g. [3]). Furthermore, recent studies point out the influence of biogenic compounds on photochemical pollution (e.g. [4]), but few investigate biogenic emission weight in air pollution control strategy selection [5].

This study is addressed to identify and quantify the role of anthropogenic and biogenic emissions on ozone concentration levels, as well as the interactions among such sources

and the pollutant masses transported inside the study domain (i.e. the boundary condition effects). The analysis has been performed by applying the factor separation technique [6]. The methodology allows to assess the impact of a single factor in a nonlinear system as well as the interaction between that factor and others processing the simulation performed by GAMES photochemical modelling system [7].

The paper is organized as follows: in the first section, an overview of the GAMES modelling system is described, focusing on the main features of each system module; in the second one, the methodological approach used to investigate the contribution of different factors involved in a nonlinear dynamic system is presented; finally, the case study aimed to evaluate the influence of boundary conditions as well as anthropogenic and natural emission sources on photochemical pollution over Northern Italy is discussed.

II. GAMES SYSTEM DESCRIPTION

In the following section, the main features of the Gas Aerosol Modelling Evaluation System (GAMES) formulation are described. GAMES, including the 3D meteorological processor CALMET [8], the emission model POEM-PM [9] and the photochemical transport model TCAM [7], allows to perform multi-phase simulations.

A. Transport and chemical model

The TCAM (Transport and Chemical Aerosol Model) is a three-dimensional Eulerian chemical transport model, that provides a exhaustive description of physical and chemical processes in the troposphere.

The evolution in the atmosphere and the chemical transformation of a specific pollutant is described by a mass balance equation. Such equation contains conservation terms (transport) and source/sink terms (emission, deposition, chemical reactions). The general mass balance equation for a generic species i , whose concentration is C_i [$g\ m^{-3}$], is then given by:

$$\frac{\partial C_i}{\partial t} = T_i + R_i + D_i + S_i \quad (1)$$

where T_i is the transport/diffusion term, R_i is the reaction term, D_i includes wet and dry deposition, S_i represents the emission term. To solve the resulting system of partial differential equations it is necessary to provide the initial and boundary conditions for each involved species.

Being the integration of 1 very time consuming and resources expensive, TCAM implements a split operator technique. This approach solves separately horizontal and vertical transport as well as the vertical phenomena of

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emissions and deposition, and the chemical processes, as follows:

$$C_i^{[n+1]} = A_{xy}A_zA_cA_cA_zA_{xy}C_i^{[n]} \quad (2)$$

where A_{xy} , A_z and A_c are the horizontal transport, the vertical phenomena and the chemistry operators respectively. At each time step, the three modules are executed for an half-time step and next they are re-executed in reverse order for the remaining half-time step. The two half applications of the chemical operator are compressed into a single operation, reducing the time computing required by the chemical integration, the most time consuming part. These three operators can be expressed by means of three differential equation systems:

$$\begin{aligned} \left[\frac{\partial C_i}{\partial t} \right]_{xy} &= -v_x \frac{\partial C_i}{\partial x} - v_y \frac{\partial C_i}{\partial y} + K_{xx} \frac{\partial^2 C_i}{\partial x^2} + K_{yy} \frac{\partial^2 C_i}{\partial y^2} \\ \left[\frac{\partial C_i}{\partial t} \right]_v &= -v_z \frac{\partial C_i}{\partial z} + \frac{\partial}{\partial z} \left(K_{zz} \frac{\partial C_i}{\partial z} \right) + S_i - D_i \\ \left[\frac{\partial C_i}{\partial t} \right]_c &= P_i(C) - L_i(C) \cdot C_i \end{aligned} \quad (3)$$

where v_x , v_y and v_z are the wind components [$m s^{-1}$], C_i is the concentration of i species [$g m^{-3}$], D_i and S_i are the deposition and emission terms, K_{xx} , K_{yy} , K_{zz} are the diffusion coefficients [$m^2 s^{-1}$] along the x , y and z direction, P_i and L_i are the production [$g m^{-3} s^{-1}$] and removal [s^{-1}] terms due to chemical reactions.

Horizontal transport and diffusion algorithms To solve the horizontal transport, both for gas and aerosol pollutants, TCAM implements a Chapeau-function-based scheme and the Forester nonlinear filter. Such algorithms, computationally efficient, allow the full mass conservation as well as the negative concentration suppression. As for the computation of horizontal diffusion coefficients K_{xx} and K_{yy} , depending on meteorological parameters, within the boundary layer, the diffusivity value is established for each of the Pasquill-Gifford stability classes [10], while above boundary layer a user-defined constant value is assigned to all species.

Vertical transport and diffusion algorithms TCAM model uses a terrain following coordinates system, characterized by vertical levels reproducing the shape of the terrain. As the turbulent diffusion generally dominates the vertical displacement of the pollutants, the numerical scheme used to solve vertical transport equation is carefully selected on the basis of the value of the vertical turbulent diffusivity coefficient K_{zz} . A fully implicit scheme is used for high K_{zz} values, while, when the atmosphere is described by stable classes, as during the night, an explicit Cranck-Nicholson scheme is used. The calculation of K_{zz} is performed taking into account meteorological parameters such as the Monin-Obukhov length, the mixing height, the friction velocity and the convective velocity.

Removal process TCAM treats dry deposition, a removal process that occurs in the atmosphere lower and concerns all the pollutants. The main factors ruling this process are: (1) the surface characteristics, as the roughness and the

composition of the surface, (2) atmospheric variables, as stability and turbulence, (3) and pollutant properties, as solubility and reactivity. The phenomenon is described by $F_i = C_i \cdot v_{di}$, where v_{di} is the deposition velocity [$m s^{-1}$], C_i is the concentration of the i specie near the terrain [$g m^{-3}$], F_i is the removed pollutant flux [$g m^{-2} s^{-1}$].

Chemical kinetics The model implements different chemical mechanisms based both on lumped molecule (SAPRC90 [11], SAPRC97 [12], COCOH97 [13]) and on lumped structure (CBIV [14]) approaches. Each chemical scheme involves a different number of treated species as well as reactions used to describe the dynamics of the accounted compounds. Table I summarizes the suitable chemical module configurations. The treatment of chemical reactions into air quality models implies the necessities to solve a set of nonlinear ordinary differential equations, according to 3. To solve the chemical system TCAM model makes use of the Implicit-Explicit Hybrid (IEH) solver [15], that divides all the species in fast and slow one, according to their reaction velocity.

B. Emission model

The emission model POEM-PM (Pollutant Emission Model for gas and Particulate Matter), designed in the framework of SATURN/EUROTRAC-2 scientific project, estimates present and alternative emission fields at both local and mesoscale level for multi-phase modelling systems, by means of an integrated top-down and bottom-up approach.

The *top-down approach* estimates emission fields disaggregating a large space-time scale pollutant activities inventory, processing surrogate variables, highly correlated with emissions and defined by means of national and local statistical sources, GIS and land use information. Such procedure is divided into four steps: (1) spatial disaggregation into municipal areas and a grid domain, (2) hourly modulation, (3) splitting of total VOC into components, and (4) chemical and granulometric splitting of PM.

The *bottom-up approach* does not involve an emission disaggregation process, but it consists of a source-oriented inquiry to obtain all the data needed to describe the emission behavior of a single source and this detailed information is then used to directly compute the emissions.

POEM-PM implements a bottom-up algorithm for the biogenic emissions, according to CORINAIR Guidebook [16]. Biogenic emissions are estimated multiplying an emission factor for a type of vegetation by a statistic giving the amount of vegetation in the country grid or grid square. The emission flux on hourly basis for biogenic VOC species i , mainly

TABLE I
TCAM CHEMICAL CONFIGURATION.

Mechanism	No. of Species	No. of Reactions
SAPRC90	54	128
SAPRC97	82	184
COCOH97	95	187
CBIV	32	81

isoprene, and due to tree species b is given by:

$$E_{i,b} = EF_{i,b} \cdot D_b \cdot \gamma_i \quad (4)$$

where $E_{i,b}$ is the hourly emission flux [$mg \ m^{-2} \ h^{-1}$], $EF_{i,b}$ is the emission factor [$mg \ g^{-1} \ h^{-1}$], D_b is the foliar biomass density [$g \ m^{-2}$], γ_i is a unitless correction factor representing the effects of temperature and solar radiation changes on emissions. As each ecosystem consists of few type of vegetation, the emission of such forest is computed adding the emission for each vegetation type estimated as in 4.

C. Meteorological model

The CALMET meteorological model includes (1) a diagnostic wind field module and (2) micrometeorological modules for overwater and overland boundary layers.

The diagnostic wind field module uses a two step approach to compute the wind fields. In the first step an initial-guess wind field is adjusted for kinematic effects of terrain, slope flows, and terrain blocking effects to produce a preliminary wind field. The second step consists of an objective analysis procedure to introduce observational data into the wind field determined in the former step to produce a final wind field.

Two boundary layer models for application to overland and overwater grid cells are accounted by CALMET model. The principal parameters needed to describe the boundary layer structure are the surface heat flux, surface momentum flux and the boundary layer height. Several additional parameters, including the friction velocity, convective velocity scale and the Monin-Obukhov length are derived from these.

III. FACTOR SEPARATION METHOD

In order to evaluate the effect of one specific factor on a given nonlinear process, a sensitive analysis can be carried out. Such analysis is performed examining the difference between two simulations, with or without the involved factor. However, this simple approach is not suitable dealing with two or more factors because of the nonlinearity of the process. To evaluate the contribution of relevant factors and the interactions among them in a particular physical process, as the pollution formation and transport in atmosphere, the factor separation method can be applied [6].

Let the field f depends on a factor c , so that:

$$f = f(c) \quad (5)$$

Factor separation technique decomposes the function $f(c)$ into a constant part that does not depend from c ($f(0)$) and a c -depending component (f'_c), as follows:

$$\begin{aligned} f'(0) &= f(0) \\ f'(c) &= f(c) - f(0) \end{aligned} \quad (6)$$

In order to solve such system, two simulations must be carried out: the former includes the factor c (f_c), and the latter does not (f_0).

Let consider three factors, namely A, B and C and let $F \doteq \{A, B, C\}$. Denoting with f_{tot} , f_0 , f_i , with $i \in F$, the simulations including all the factors, none of the factors and

factor i only respectively, the pure contribution of factor i , denoted with f'_i , can be isolated as follows:

$$\begin{aligned} f'_0 &= f_0 \\ f'_i &= f_i - f_0, \quad i \in F \end{aligned} \quad (7)$$

Note that the term f'_i represents the fraction of the considered process completely ascribed to factor i , while f_0 is the part independent from i .

The double mutual interaction among the factors i and j is given by:

$$f'_{ij} = f_{ij} - (f_i + f_j) + f_0 \quad (8)$$

$$i, j \in F, \quad i \neq j \quad (9)$$

The triple interaction is expressed as:

$$f'_{tot} = f_{tot} + \sum_{i \in F} f_i - \sum_{\substack{i, j \in F \\ i \neq j}} f_{ij} - f_0 \quad (10)$$

In this way, the run including all the involved factors in term of pure contributions is obtained by the following equation:

$$f_{tot} = f'_{tot} + \sum_{i \in F} f'_i + \sum_{\substack{i, j \in F \\ j \neq i}} f'_{ij} + f'_0 \quad (11)$$

As introduced by [17], the total impact (TI) of a specific accounted factor i is defined as the sum of all pure contribution and synergic terms in which i appears, as follows:

$$\begin{aligned} TI_i &= f'_{tot} + f'_i + \sum_{\substack{i, j \in F \\ j \neq i}} f'_{ij} \\ &= f_{tot} - f_{jk} \\ &\quad j, k \in F, \quad j \neq k \neq i \end{aligned} \quad (12)$$

IV. CASE STUDY

This study focuses on a Northern Italy region. Bounded north by Alps, it covers a large part of the Po Valley, presenting an agricultural area in the southern part, along the Po River, and a densely urbanized area in the central part. In Northern Italy two crucial aspects have to be accounted in selecting control strategies. (1) The critical anthropogenic emissions, the frequent stagnating meteorological conditions and the Mediterranean solar radiation regularly cause high ozone level concentrations. (2) Moreover, the domain is ruled by both NOx-VOC sensitive regimes. In such a complex domain, several experimental campaigns (e.g. [18]) as well as modelling studies (e.g. [19],[20]) have been carried out to investigate photochemical pollution formation processes.

In this work, GAMES system has been used to carry out homogenous gas-phase simulations implementing SAPRC90 mechanism, and the period April-September 1996 has been selected as the reference case. The selected area (Fig. 1) is $240 \times 232 \text{ km}^2$ wide and it has been subdivided with a horizontal grid resolution of $4 \times 4 \text{ km}^2$, with 11 vertical variable layers (20, 45, 80, 130, 230, 400, 650, 1000, 1700, 2800 and 3900 m a.s.l.).

GAMES input fields have been provided as follows. *Emission fields* The road transport, agriculture and biogenic emissions have been estimated by a bottom-up approach, while emissions of the other sectors have been disaggregated, modulated and splitted on the basis of the Italian CORINAIR database. Fig. 2 and Fig. 3 show the biogenic emission weight [%] to total NO_x and NMVOC emissions over the whole domain respectively. Both maps highlight that in the N-O part, in correspondence of Switzerland domain, biogenic sources play a dominant role. Apart from Switzerland region, biogenic contributions to NO_x total emissions rise up to 60%, localized in limited areas. For NMVOC emissions, the higher biogenic emissions are evidenced in Alpine region and in correspondence of the Po river.

The *meteorological fields* have been provided by CALMET model starting from three data sets: (1) the land-use provides parameters for turbulence and deposition modules; (2) the radio-sounding measurements and ECMWF model 6-hourly simulations have been processed to build vertical wind structure and temperature fields; (3) the hourly ground level data measured in 31 stations account for local effects.

Boundary conditions have been defined processing the outputs of EMEP model [21]. It is a one-layer Lagrangian trajectory model that covers the whole Europe, using a polar stereographic grid with a resolution of 150×150 km² at 60°N. It provides boundary layer mean concentrations at 0, 6, 12 and 18 GMT.

The performance assessment of the integrated modelling system as for the base case simulation over the selected domain is described in detail in [20], [22], [23], including the estimate of the indicators required in the EC Directive 2002/3 [24] as well as the recommended US EPA indexes [25]. Fig. 4 shows the ozone mean value pattern computed for the summer period 1996. The geographical distribution of ozone mean value computed for the summer period 1996 points out a general increasing gradient from the central part of the domain to North and South, so that lowest values are found in main urban areas, namely Milan metropolitan area,

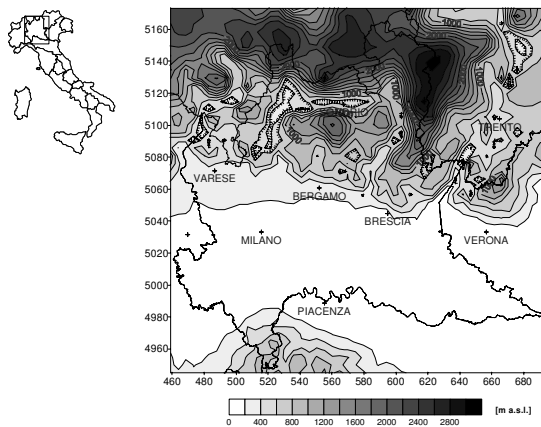


Fig. 1. The selected domain topography with Lombardia Region boundaries and main cities.

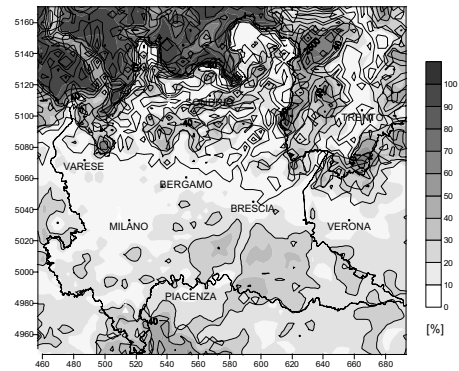


Fig. 2. Biogenic contribution [%] to NO_x total emissions.

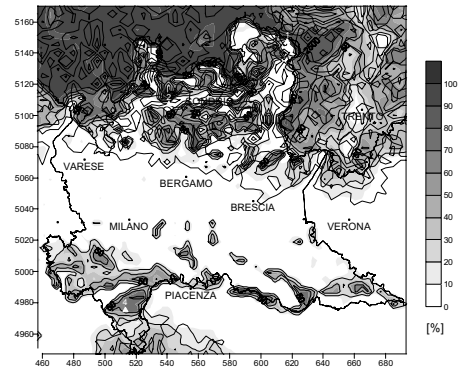


Fig. 3. Biogenic contribution [%] to NMVOC total emission.

V. RESULTS AND DISCUSSION

According to factor separation technique, three factors have been accounted: (A) the anthropogenic emissions, (B) the biogenic ones, and (C) the boundary conditions. Because of three factors are used, eight simulations must be carried out (Table II).

A first evaluation of the weight of boundary conditions and emission sources is carried out analyzing the pure impact of each factor on the ozone concentration level. Fig. 5, Fig. 6 and Fig. 7 show the pure anthropogenic (f'_A), the pure biogenic (f'_B) and the pure boundary condition contribution (f'_C) respectively. Such maps pointed out that the contribution of each factor is always positive, with higher values due to anthropogenic emissions and boundary conditions. Further,

TABLE II
SUMMARY OF THE SIMULATION INPUTS.

Simulation	Anthropogenic emissions	Biogenic emissions	Boundary conditions
f_o	-	-	-
f'_A	✓	-	-
f'_B	-	✓	-
f'_C	-	-	✓
f'_{AB}	✓	✓	-
f'_{AC}	✓	-	✓
f'_{BC}	-	✓	✓
f'_{tot}	✓	✓	✓

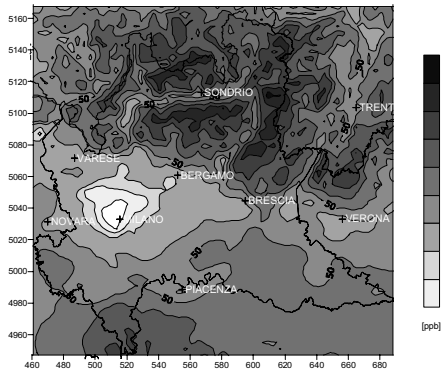


Fig. 4. Ground level ozone mean value distribution, computed over the 6-months period [ppb].

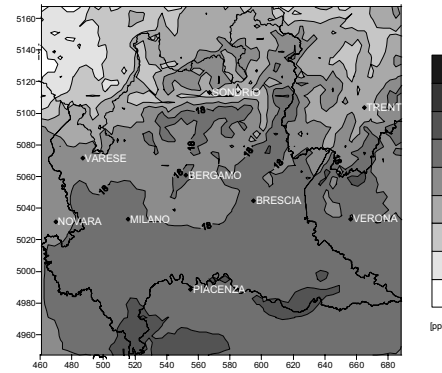


Fig. 6. Pure biogenic impact f_B' [ppb] at ground level.

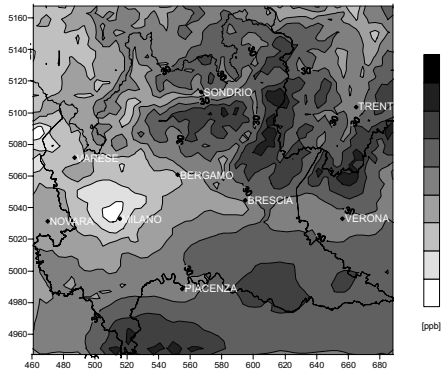


Fig. 5. Pure anthropogenic impact f_A' [ppb] at ground level.

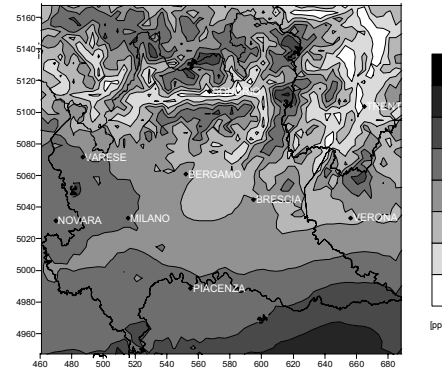


Fig. 7. Pure boundary condition impact f_C' [ppb] at ground level.

the lower bound of boundary condition pure contribution is higher (24 ppb) than the emission source ones (about 10 ppb).

As the analysis performed by means of the pure contribution does not account the nonlinearity among the factors, to evaluate the complete contribution of each factor on the mean ozone concentrations the total impact has computed according to 12. Fig. 8, Fig. 9 and Fig. 10 show the total anthropogenic impact ($TI_A = f_{tot} - f_{BC}$), the total biogenic impact ($TI_B = f_{tot} - f_{AC}$) and the total boundary condition impact ($TI_C = f_{tot} - f_{AB}$) respectively.

Focusing on TI_A , the impact ranges between -24 ppb to 16 ppb. Negative values, i.e. ozone reduction, are estimated over high NOx anthropogenic emission area, as the presence of high anthropogenic emission produces a reduction of ozone concentration, caused by VOC-limited regime characterizing such area [20]. This behavior is more evident for the Milan metropolitan area. Maximum values of TI_A are reached in northern part of domain, that is NOx-limited [20].

The biogenic contribution, mostly positive, presents the maximum impact in the correspondence of the high biogenic emission areas, i.e. the Alpine region and along the Po river, in domain southern part.

The maps showing the boundary condition total impact highlight that: (1) positive contributions are quite similar to the anthropogenic and biogenic ones; (2) TI_C is null in correspondence of Milan; (3) the urbanized area between

Milan and Verona is characterized by a modest impact (only few ppbs); (4) the highest impact is found in the NW part of the domain, in correspondence of Switzerland area. Thus, total boundary condition effects in the area mainly characterized by elevated local anthropic emissions, as the Po Valley, are quite modest, while they mainly influence areas with moderate local emissions (Fig. 2, Fig. 3).

VI. CONCLUSIONS

GAMES modelling system has been applied to a seasonal simulation (April-September 1996) over Lombardia Region (Northern Italy) in order to assess the influence of boundary conditions and emissions on tropospheric ozone formation and accumulation processes.

This analysis represents a useful tool for regional authorities interested in developing plans for ozone reduction, allowing to deduce the following evidences. (1) The biogenic impact is positive in all the domain, meaning that this contribute cannot be neglected in planning an air quality strategy. (2) In the northern part of the domain, an area characterized by low precursor emissions, ozone is equally produced by transboundary processes, by mountain-valley breeze transport from urban area and by biogenic emissions. (3) Control strategies based on local anthropogenic emission reduction measures have limited expected effectiveness in domain northern part. This result agrees the analysis performed by means of a different approach, i.e. a sensitivity assessment on ozone precursor emissions [20].

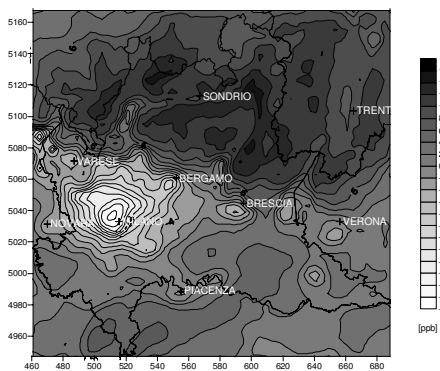


Fig. 8. Total anthropogenic impact TIA [ppb] at ground level.

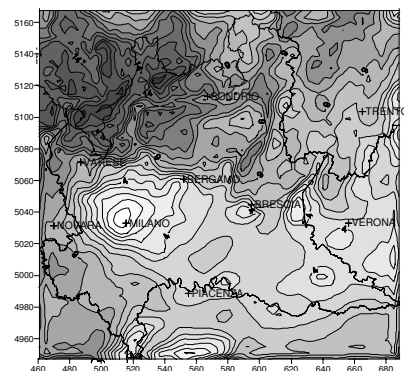


Fig. 10. Total boundary condition impact TIC [ppb] at ground level.

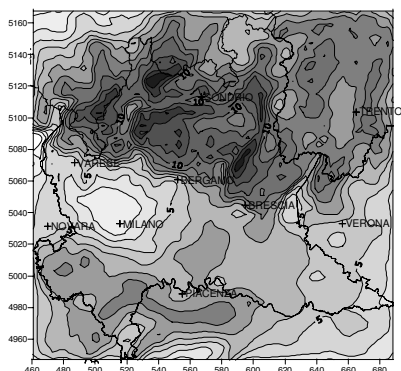


Fig. 9. Total biogenic impact TIB [ppb] at ground level.

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