A neural emission-receptor model for ozone reduction planning

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Abstract: Ground level ozone pollution is a complex phenomenon heavily affecting industrialized and populated areas. Ozone is produced by a series of photochemical reactions, activated by the emissions of nitrogen oxides and volatile organic compounds and may reach maximum concentrations at kilometers of distance form the precursors sources, depending on the meteorological conditions. Models to compute ozone concentrations are equally complex and cannot be directly used to optimize emission reduction policies. For this reason, a neural network has been trained on the results of a photochemical model (CALGRID) to represent the emission-receptor relationships in critical conditions. Such a network in then entered in an optimization problem that determines the least cost alternatives to obtain a given air quality standard. The decision variables of the problem are the emission reductions of ozone precursors in each industrial sector. These reductions are in turn the result of the application of a number of technologies, whose costs and performances are known. The approach has been used to estimate the optimal reduction alternatives for a region in Northern Italy and showed that a consistent improvement of air quality can be attained with moderate investments, provided that these are concentrated in some sectors, such as road transport and industrial solvents, that have a major impact on ozone dynamics.

Keywords: neural networks, ozone ground pollution, Lombardy region

1 INTRODUCTION

High ozone concentrations at the tropospheric level are a major concern in air pollution studies because of their impact on human health and agricultural crops and forests. High ozone concentrations have been observed since the 1970s in the United States and in Europe; since high temperatures result in a much quicker ozone formation, cities with warm climates experience usually severe ozone problems, as observed for Milan (Silibello et al. [2000]). In particular, Lombardy region, located in Northern Italy, experiences heavy photochemical pollution almost every summer.

In order to be effective in ozone concentration lowering, control policies should be focused on ozone precursors reduction, i.e. NO_x and volatile organic compounds (VOC). As well known, ozone formation is controlled either from NO_x or VOC, depending on the ratio between their concentrations: when NO_x/VOC is "low", the rate of ozone formation increases with NO_x and changes due to increased *VOC* are negligible ($NO_x - sensitive$ regime). At high ratio levels, ozone concentration decreases increasing NO_x and increases increasing VOC (*VOC-limited* or $NO_x - saturated$ regime). Hence, chemical sensitivity analysis plays a key role in developing successful ozone reduction policies: as an example, NO_x reductions will be effective only in $NO_x - sensitive$ regimes (Sillman [1999]). According to *CORINAIR* classification, emission sources can be grouped in 11 sectors; the policy design has hence to take into account that precursors emission reduction will have different costs depending on the considered emission sector. An evaluation of the marginal costs for precursors emission reduction in each sector for different European countries can be found in Klimont et al. [2000].

The aim of this paper is to tackle the problem of air quality optimization for Lombardy region, determining the costs requested for a given ozone level reduction; a reduction policy is hence defined by a set of reduction rates of ozone precursors, which refer to different emission sectors.

From a modelling point of view, the relationship between ozone and its precursors is captured through a neural network, thus exploiting the network ability in recognizing highly non linear relationships. Indeed, artificial neural networks have been showed to be able to well represent air pollution phenomena in many previous works (see for instances Gardner and Dorling [1998]). Photochemical 3-dimensional models such as CALGRID (Yamartino et al. [1992]), while providing a very detailed simulation of the complex photochemical processes, cannot be used in an optimization task because of the computational effort involved. On the contrary, neural network can be successfully used in such application because of their computational speed.

2 Lombardy case study

Lombardy Region, which has an overall area of about $24000 km^2$, compraises a plain part (47%) located in the Po Valley, a hilly (12%) and a mountainous district (41%). The plain part, heavily industrialized and populated, frequently presents stagnating meteorological conditions which cause high ozone levels during summer. National and Regional laws have fixed attention and alarm levels for ozone concentration based on hourly averages, but recently (DM 16/5/96) a limit of $110 \mu g/m^3$ on the 8-hour average has been established, following the directories of the World Health Organization on the protection of human health.

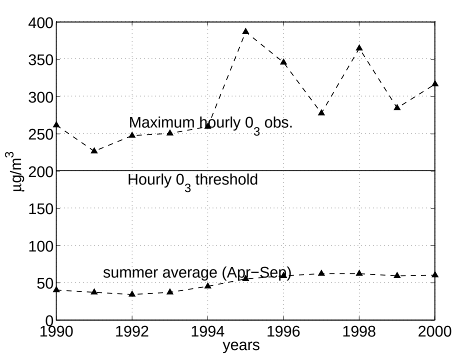

Figure 1: Ozone trends for the metropolitan area of Milan.

Considering the air pollution situation in the region, it should be noticed that, during the 50's and 60's, the atmosphere showed mainly a reducing behavior, due to high SO_2 concentrations. The development of private transport during the 80's and the consequent increase in mobile sources emissions resulted in a significant increase in NO_x and CO values. Starting from the early 90's however, SO_2 has begun to decrease significantly thanks to the intensive use of heating oils with lower sulphur contents, while the catalytic converters adoption (forced by the national law) has allowed to lower *CO* and NO_x levels. The reduced SO_2, NO_x and *CO* concentrations have caused a more oxidant behavior of the atmosphere and, as a consequence, O_3 levels have begun to increase from the early 90's. Ozone trend in the metropolitan area of Milan over the last decade (MilanMunicipality [2000]) is shown in Fig.1; yearly average is computed with regard only to the warmer period of the year, namely between April 1st and September 30th. In the last years, ozone exceeded human health threshold for daily average at almost all (96%) measurement stations, thus claiming for careful design of primary pollutants reduction policies.

Figure 2: Sites with elevation lower (light) or higher (dark) than 300m within Lombardy region.

Photochemical sensitivity of the region has been analyzed simulating different precursors reduction scenarios, assumed uniform on the whole domain, and then evaluating changes in ozone levels (Gabusi and Finzi [2000]) or in specific species ratios, such as O_3/NO_x (Silibello et al. [2000]). The analysis individuated the *VOC sensitive* area on the plain part, which experiences high anthropogenic emission rates, and on the pre-alpine zone impacted by urban plumes. In contrast, one could suppose the *mountain district* to have a NO_x *sensitive* chemistry, since it is characterized by more aged air masses and high biogenic emission rates, according to Sillman [1999]. Photochemical simulations show, as expected, that emission control strategies focused on VOC reduction are not effective for this part of the domain; more noticeable, NO_x *reductions even in the order of 30% produce negligible* *effects on ozone levels*. High ozone levels measured in this part of the domain need hence further investigation in order to be fully understood and to allow the design of an effective control policy. Since ozone concentrations in the mountain district seems to be insensitive to the precursors reduction, we will focus our study only on the plain part of the domain, i.e. with an elevation above sea level lower than 300m (see Fig. 2). Estimated *VOC* emissions in the whole region presently amount to about 1110 ton/day; 80% of them origins from solvent use (470 ton/day) and road transports (408 ton/day); less influential are waste treatment (110 kg/day), fossil fuels distribution (50 ton/day) and production processes without combustion (23 ton/day).

2.1 State of the art

A simplified quadratic *source-receptor* model for ground level ozone can be found in (Schopp et al. [1999]); the model predicts daily ozone concentration at each receptor taking into account *VOC* and NO_x emission rates at the sources. This simplified description of the source-receptor relationship can be used within an integrated assessment model, thus allowing for a systematic cost-effectiveness analysis. It should be noted that more complex models, which contain a high degree of detail of chemical and meteorological processes, can not be employed in optimization analysis because of their computational requirements.

Following in part this approach, Guariso [2000] formulated for Lombardy region a two objectives optimization problem, namely ozone level reductions and minimization of related costs. For this purpose, he gridded the domain on *4 km * 4km* cells and simulated several precursors reduction scenarios using CALGRID. Then, on the base of the simulations results, he identified on each cell of the domain, a quadratic relationship between NO_x and VOC emissions at the sources and ozone concentrations at the receptor. The optimization problem, formulated at regional scale, exploited such functions in order to evaluate the effectiveness of given reduction policies. As a result, the computed Pareto boundary showed a maximum curvature for a reduction degree of about 40% of maximum feasible reduction, while costs were about just 15% of the costs sustained in the case of maximum feasible reductions.

Aim of this paper is to solve a similar optimization problems, improving some modelling issues. The calibration of a quadratic relationship in each cell of the domain results in a quite low ratio data/parameter, thus possibly forcing the parameters to capture also the noise contained in the data. Instead of calibrating several hundreds quadratic relationships, we choose to take into account the photochemical behavior dissimetry by selecting the *VOC-sensitive* part of the domain, and then training a neural network on this partition. Such network use the same parameters set for all the cell belonging to the partition, thus significantly improving the data/parameters ratio. This results in a better generalization of the network with respect to the local quadratic models, and makes the analysis of the reduction policies more reliable.

3 NEURAL SOURCE-RECEPTOR MODEL

We trained a neural network model aimed at capturing the relationship $NO_x - VOC - O_3$ for the *plain* part of the domain. The *maximum 8-hours average ozone concentration* during the current day on each cell, i.e. on each receptor, was chosen as process variable. Let us define, for a given receptor of coordinates (i, j) on the gridded domain, the neighboring set S_{ij} consisting of the 8 outer cells in the square centered on position (i, j). In order to capture the relationships between ozone and its precursors, we use the following input variables:

- overall NO_x and *VOC* emissions during the previous day;
- initial NO_x and *VOC* concentrations;
- elevation of the cell above sea level.

Since input variables are evaluated at each cell belonging to S_{ij}, the resulting input set is compound by $45 = 5 * 9$ variables. In order to lower the input dimensionality, we removed input variables which shows too low variances through the *Principal Component Analysis* (PCA). Thus, the input set reduces to 22 variables. We focused on a one hidden-layer network with hyperbolic tangent as transfer function for the hidden nodes and a linear output node, since such architecture has been widely shown as suitable in this kind of application (Gardner and Dorling [1998]).

The available dataset contains CALGRID outputs for five different emission scenarios, simulated on the meteorological conditions of 5-7 June 1996, which has been a major pollution episode in Lombardy and entire Europe during the last years. The simulated scenarios assume a 35%, 50%, 60% uniform reduction in just *VOC* or NO_x, or in both the pollutants at the same time. Such reductions are computed with respect to the *base* case, namely the estimated emission patterns for June 1996.

The whole dataset has been divided into a *training set*, used to estimate the parameters of the neural networks, a *validation set*, used to evaluate the generalization capability of the networks, and finally a *testing set*. In order to avoid overfitting, we exploited the *early stopping* technique, evaluating at each iteration the objective function (actually the *sum of squared errors*) on the training and on the validation set. According to Bishop [1995] the training is stopped once the error function on the validation set begins to increase. We analyzed exhaustively combinations of PCA threshold and number of neurons n in the hidden layer, by trial and error; each configuration has been trained several times and by means of different training algorithm, such as standard backpropagation and Levenberg-Marquardt. Finally, the network has been evaluated on the testing set, *not involved in any training phase*. The best performances were obtained using

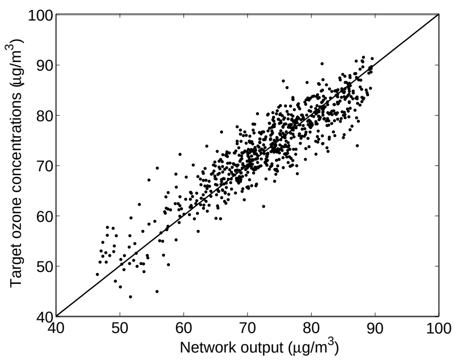

Figure 3: Comparison between network targets and outputs. *testing set*

Levenberg-Marquardt as training algorithm, which is in fact recognized as very suitable in the estimate of small and medium size networks (Bishop [1995]); with regard to number of nodes in the hidden layer we obtained the best results with $n = 20$. A comparison between networks outputs and targets, i.e. CALGRID outputs, is showed in Fig.3, while some performances indicators are collected in Tab.1.

If we consider the complexity of the problem, the model provides satisfactory performances: high correlations between CALGRID simulations and network outputs, acceptable level of over and under estimation. The ANN modelling approach results in 480 parameters to be estimated, compared to the about 6000 for the local quadratic approach; one can then easily understand the improvement in the generalization capability of the model.

	Training	*Testing*
ρ	.961	.912
Mue	-17.72%	-28.87%
Moe	23.18%	38.73%

Table 1: Network performances: correlation ρ between network target and outputs; maximum underestimate Mue and maximum overestimate error Moe.

3.1 Determination of the Pareto boundary

The purpose of the whole research is to find out a set of efficient solutions which, for a given cost level, shows the maximum feasible ozone reduction. We have hence to solve a multi-objective optimization problem.

In the ANN modelling phase, aimed at capturing the physical relationship between ozone and its precursors, we did not care about emission sources; however, the optimization phase, aimed at providing a different reduction rate for each sector, requires to group emissions according to the subdivision previously explained.

With regard to the spatial aggregation of the decision variables, the work of Guariso [2000] assumed a uniform reduction rates for each sector on the whole domain. Although one could in principle configure a different reduction rate for every sector on every cell, it should be noticed that in the real world it is in general not possible to take so *fine-grained* decision. Consistently with the ANN modelling phase, we assumed uniform reduction rates for each emission sector on the whole plain partition. This solution has also the advantage to preserve the computational feasibility of the problem, since it leads to a overall optimization problem which contains only 5 decision variables. In fact, we did not take into account emission sectors such as power plants, combustion, off road transports, which play a minor role; moreover also biogenic emission have not been treated in the analysis, since such emissions can be modified only changing the land use.

Therefore we focused our analysis only on the most influential *VOC* emission sectors: *production processes without combustion* (sector 4), *extraction and* *distribution of fossil fuels* (sector 5), *solvent use* (sector 6), *road transport* (sector 7), *waste treatment and disposal* (sector 9). Let us denote with s the *VOC* emission sector, with r_s the *VOC* reduction rate for the *s*-th sector and with $E_s^{i,j}$ the *VOC* emission of cell *(i,j)* for the *s*-th sector in the base scenario. The optimization problem can be formulated as:

$$\min(costs) = \min_r \sum_s [r_s * E_s^{i,j} * c_s(r_s)] \qquad (1)$$

$$\min(pollution) = \min_r \sum_{i,j} I_{i,j}(r_s) \qquad (2)$$

constrained by

$$0 \leq r_s \leq R_s \qquad (3)$$

where $I_{ij}(r_s)$ is the value of the air quality indicator for cell *(i,j)*, i.e. the output of the previously trained ANN, and R_s is the maximum *feasible VOC* reduction for sector *s* with the set of technologies considered. Unitary emissions reduction costs $c_s(r_s)$ and their dependence on reduction rates r_s have been evaluated according to the estimates published in Klimont et al. [2000] for the Italian situation.

4 RESULTS

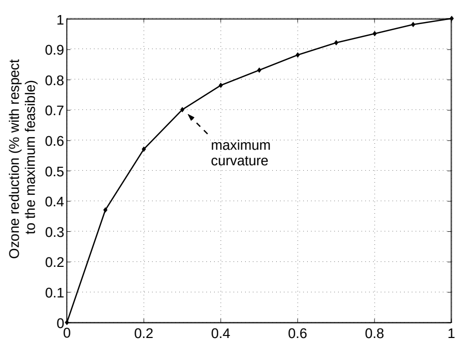

Figure 4: Pareto boundary

The problem was solved using the constraint method: the optimization of air quality has been performed at fixed cost levels. The obtained *Pareto-boundary* is showed in fig. 4.

As one can notice, the pollution index decreases rapidly till 70% of the feasible reduction: this happens at a cost level equal to 30% of the costs for the maximum feasible reduction. One can hence suggest a 60%-80% pollution reduction level, whose associated costs are between 20% and 40% of the maximum costs; in fact it is hardly acceptable to operate beyond the 80% of pollution reduction, because of the high costs involved. We will analyze in greater detail the solution (70% pollution, 30% costs), which one would choose if operating according to the *maximum curvature criterion.*

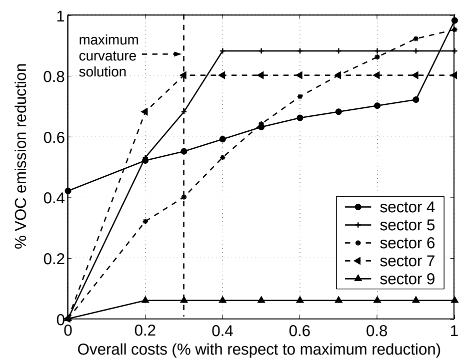

Figure 5: Reduction rates on the different sectors for a given overall cost level.

VOC reduction rates r_s corresponding to a given cost level in the Pareto boundary are showed in Figure 5. With regard to the most influential sectors one can notice that:

- sector 7 (road transport), which has a feasible reduction rate of 80%, is severely reduced even in low cost situations. The proposed solution will decrease it as much as possible;
- sector 6 (solvent use), which has a feasible reduction rate of 95%, has high reduction rates only for high costs situations. In the proposed solution it is lowered by 40%.

With regard to the remaining sectors: sector 9 (*waste treatment and disposal*) has a very low feasible reduction rate (6%), which is fully implemented in the proposed solution; sector 5 (*fossil fuels*), which has a feasible reduction rate of 88%, is reduced by about 68% in the proposed solution; sector 4 (*production processes without combustion*), which has a feasible reduction rate of 98% is reduced by about 55%.

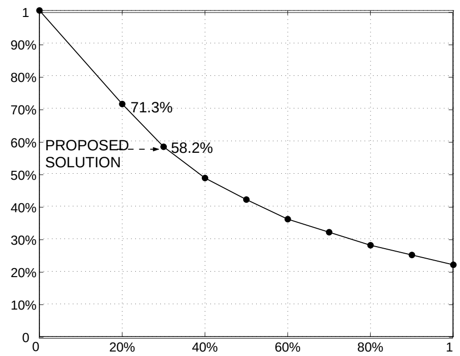

Figure 6: VOC emission reduction compared to their associated costs.

For a different point of view, it could be of interest to examine the relationship between intervention costs and VOC emissions, shown in Fig.6; as one can notice, costs are almost linear till 50% VOC reduction, while marginal cost increases rapidly for higher reductions. The overall relationship between VOC reduction costs and ozone level improvement can be thus thought as composed by two different steps: the first one, which joins VOC reduction levels to the requested costs, and a second one, which maps VOC emissions to ozone levels, namely the Pareto boundary shown in Fig.4. The proposed solution involves a 30% costs level and a 42% emission reduction; it is noticeable that such emission reduction results in a much higher (70%) ozone indicator improvement.

5 CONCLUSIONS

The purpose of this paper has been to investigate the main priorities of emission reductions in order to low tropospheric ozone pollution in Lombardy region. According to the photochemical characterization of the domain, we focused our analysis on VOC emission reduction in the plain part of the Region; a multi-objective (*cost, pollution*) optimization problem has been solved exploiting the computational speed of the network trained on CALGRID output. Results show that noticeable improvements in ozone level are reachable even through moderate investments, provided that these are concentrated in some sectors, such as road transport and industrial solvents.

The whole analysis can be further developed detecting, from a photochemical point of view, a higher number of homogeneous areas in order to provide more specialized reduction patterns. Furthermore, a more detailed evaluation of the emission reduction costs for the specific plants of the region would be needed to check which of the considered technologies can be actually applied in the area. With regard to the mountain part of the domain, a more detailed photochemical sensitivity analysis is needed before designing precursors reduction policies.

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