APPLICATION OF A LAGRANGIAN PARTICLE MODEL TO THE SOURCE APPORTIONMENT FOR PRIMARY MACROPOLLUTANTS IN TARANTO AREA (SOUTH ITALY)

A. Morabito[1], R. Giua[1], A. Tanzarella[1], S. Spagnolo[1], T. Pastore[1], M. Bevere[1], E. Valentini[1], V. La Ghezza[1] , G. Assennato[1], G. Tinarelli[2], G. Brusasca[2], G. De Gennaro[3]

[1]Regional Environmental Protection Agency, Bari, Italy
[2]ARIANET, Milano, Italy
[3]Chemistry Department, University of Bari, Italy

Abstract: Dispersion models based on emission inventories and meteorological fields are the primary tool of control agencies to support air quality assessment and source apportionment in complex industrial areas. In this work, a modelling system has been applied to estimate the annual contribution to the total concentrations of different pollutant sources in Taranto, one of the most industrialized areas in Italy, where typical urban emissions are superimposed on industrial ones located in proximity of the city boundary. Main industrial activities consist of an integrated steel plant (one of the largest in Europe) and an oil refinery, together with other smaller industrial facilities which use the Taranto harbour to unload primary goods and to deliver final products.
Modelling system includes the meteorological models SWIFT-SURFPRO and the Lagrangian particle dispersion model SPRAY. The air emissions inventory is partially established using local activity indicators and emission factors. The resolution level of the data is the municipality. In particular, in this study industrial sources (point sources and fugitive), traffic, domestic heating and harbour emissions have been taken into account.
The meteorology in the studied area was reconstructed by the SWIFT model from the tridimensional meteorological products supplied, for the year 2007, by the national MINNI project.
The annual simulation led to the identification of the main emitting sources and to the source-apportionment of primary pollutants at selected receptor sites, belonging to the air quality monitoring network. Industrial activities were found to be the principal contributor to SO2 emissions. Industry and traffic emissions were, for the most part, responsible for NOx simulated concentrations, while primary PM10 and PM2.5 simulated concentrations appeared to be linked to industrial emissions. Finally, in order to demonstrate the level of representativeness of the system used in this study, the model predictions were compared with measured air quality data.

Key words: *impact of industrial plants, coupling of meteorological and dispersion model, source apportionment*

INTRODUCTION

A useful application of a Lagrangian modelling system is the assessment of the source contributions to primary pollutant concentrations in a complex meteorological area, where industrial emissions are particularly relevant.
The aim of this study has been to evaluate the relative role of different sources to the concentrations of macropollutants (SO2, NOx, PM10, PM 2.5 and C6H6) in Taranto city, located in the south-eastern part of Italy.

Due to its coastal position, Taranto is subjected to a complex sea-breeze phenomena that influences the boundary layer dynamics and the atmospheric stratification, which plays an important role in the dispersion and accumulation of air pollution. Moreover, Taranto, according to the European Pollutant Release and Transport Register (E-PRTR, 2012), is among the most highly industrialized cities in Italy, where shipping, industries and urban activities coexist in close range. The Taranto area is identified in Italy to be at high risk of environmental crisis. The air quality monitoring, carried out by the Regional Environmental Protection Agency (ARPA), at stations close to the industrial area, revealed during 2007-2008 a number of PM10 daily exceedances greater than that allowed by the air quality directive (2008/50/EC).

EMISSION DATA AND AIR QUALITY MONITORING STATIONS

The emissions of primary compounds such as NO_x, SO_2, PM_{10}, $PM_{2.5}$ and C_6H_6 from industry, traffic, domestic heating and the harbour were derived from the 2007 regional atmospheric emission inventory (ARPA Puglia, 2011), built up on the basis of INEMAR (INventario di EMissioni in Aria). INEMAR is mostly based on internationally accepted estimation methodologies (EEA-CORINAIR, 2009; JRC/BREF, 2012; EPA, 1997; IPCC Guidelines, 2006) and/or on the scientific literature. The smallest spatial resolved scale is represented by the municipality but the inventory used in this study has been appropriately detailed and integrated by specific data and information necessary to better characterize the spatial and temporal emissions.

The main industrial activities considered are an integrated steelworks, a cement factory and an oil refinery (see Fig.1). In particular 238 point (with height from 10 to 250m), 16 areal and 3 linear sources have been taken into account. The areal sources include *warm* emissions produced by the steelworks (coke ovens, sintering plant, blast furnace and steel plants) and *cold* emissions generated by the petroleum processing products, the wind

erosion in mining parks, the handling on the conveyors (located away from the urbanized area) and materials transport. The linear sources are explicitly the conveyors located closer to the urbanized area.

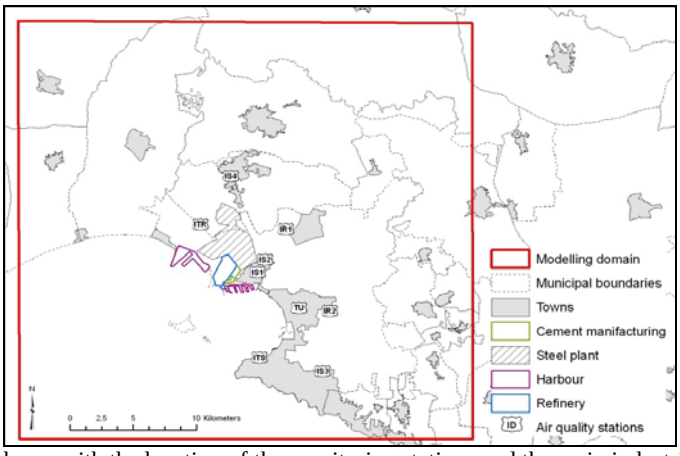

Figure 1. Map of the studied area with the location of the monitoring stations and the main industrial, harbour and urban areas.

Table 1 shows the yearly emissions for simulated emission sectors.

Table 1. Total yearly emissions for industrial sources, road traffic, residential heating and harbour activities.

Emission sources	PM10 (Mg/year)	PM2.5 (Mg/year)	C6H6 (Mg/year)	NOx (Mg/year)	SO2 (Mg/year)
Industrial activities	6460	4376	287	20439	24285
Road transport	317	274	66	3634	25
Harbour	403	381	5	3384	3041
Domestic heating	232	224	43	222	48

Nine air quality monitoring stations (see Fig. 1), managed by the Regional Environmental Protection Agency (ARPA), are located in the modelling domain. These stations daily report air quality data and are distinguished according to prevalent emission source and area type, as defined by conventional classification established by the Italian regulation: six are industrial stations, divided among suburban and rural areas (i.e. IS or IR), two are industrial/traffic stations (one suburban and one rural, i.e. ITR and ITS), and one is a traffic urban site (TU, via Adige). Table 2 summarizes the station locations and their characteristics.

Table 2 Location and characteristics of the monitoring stations.

Stations	X-UTM (km)	Y-UTM (km)	type	Monitored parameters
IS1- Via Machiavelli	688.64	4484.37	Industrial suburban	CO,C6H6,PM10,NO2,O3,SO2
IS2 - Via Archimede	689.24	4485.03	Industrial suburban	CO,PM10,NO2,SO2
IS3 – Talsano	693.78	4475.99	Industrial suburban	PM10,NO2,SO2
IS4 – Statte	686.53	4492.53	Industrial suburban	PM10,NO2,SO2
IR1 - Paolo VI	690.89	4488.02	Industrial rural	PM10,NO2,SO2
IR2 - Casa Circond.	694.36	4481.09	Industrial rural	PM10,NO2,SO2
ITR - SS7 Wind	684.11	4488.42	Industrial/traffic rural	CO,C6H6,PM10,NO2,SO2
ITS - San Vito	688.78	4477.12	Industrial/traffic suburban	NO2,SO2
TU - Via Adige	691.92	4481.34	Traffic urban	NO2,SO2

MODELLING SYSTEM SETUP

The modelling system included three models: the SWIFT meteorological model, the SURFPRO turbulence pre-processor and the SPRAY Lagrangian particles dispersion model (Tinarelli et al, 2000; Gariazzo et al., 2007). SPRAY is a 3D model particularly suited to provide an accurate local distribution of the primary pollutants in the atmosphere in non-homogeneus and non-stationary conditions.

The simulation grid was centred on the industrial area and covered a surface of 1225 km^2, with an horizontal spatial resolution of 500m x 500 m and 71 x 71 grid points. The vertical grid was divided into 15 levels with different thicknesses, starting at the surface and gradually reaching a maximum of 5000 m at the top.

The meteorology in the studied area was reconstructed by SWIFT model from the tridimensional meteorological products supplied, for the year 2007, by the MINNI project (www.minni.org, Zanini, 2009).

The warm areal emissions were not considered to be released at the ground level, but subjected to plume rise. In particular for the coke oven specific algorithms developed by EPA (EPA, 2003) were considered.
In order to provide the appropriate time resolution, the emissions from the harbour, traffic, domestic heating and industrial point sources were subjected to temporal modulation on the basis of published profiles available in official records, such as annual hours of work or local data. All the areal industrial emissions were considered constant with the exception of the dust emissions generated by the wind erosion of mineral deposits. These emissions were calculated hourly as a function of wind gusts magnitude on mineral storage piles (EPA, 1997).

RESULTS AND DISCUSSION

As example of the model results, the annual average concentration maps for NOx (produced by the all emission sectors simulated) and for primary PM10 (produced by the industrial sources), predicted by the SPRAY model, have been shown in the figure 2.

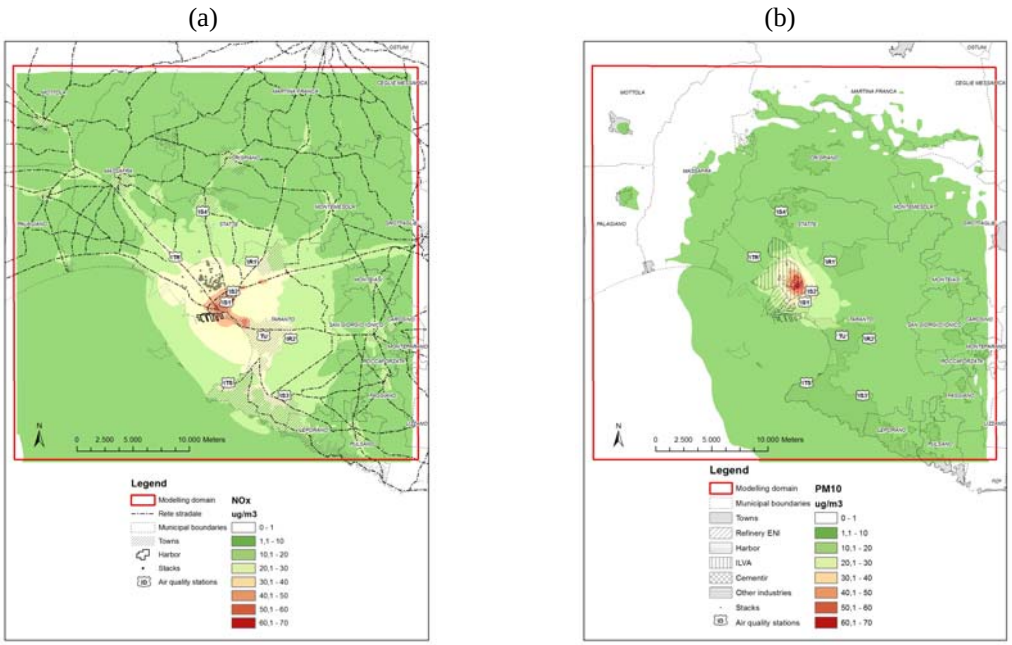

Figure 2. Annual average concentration map for (a) NOx (produced by the all emission sectors simulated) and (b) PM10 (produced by the industrial sources), predicted by the SPRAY model.

To better understand the influence of different emission sources on pollutant concentrations in different parts of the studied area, a quantitative source apportionment has been performed and shown in Table 3.

Table 3. SPRAY modelled source contributions at the monitoring stations.

Stations	**IS1**	**IS2**	**IS3**	**IS4**	**IR1**	**IR2**	**ITR**	**ITS**	**TU**
NOx									
Total annual predicted concentration (µg m^{-3})	**49.5**	**44.1**	**21.6**	**13.1**	**21.5**	**17.1**	**25.7**	**18.3**	**31.8**
Industrial activities (%)	46	49	36	47	47	49	23	43	37
Road Transport (%)	21	30	47	39	41	29	61	29	39
Harbour (%)	32	20	15	12	12	19	16	27	21
Domestic heating (%)	1	1	1	2	1	4	0	1	4
SO2									
Total annual predicted concentration (µg m^{-3})	**36.7**	**29.8**	**11.6**	**8.5**	**13.4**	**11.4**	**9.2**	**13.1**	**18.8**
Industrial activities (%)	61	73	73	82	82	73	58	66	66
Road Transport (%)	0	0	1	0	0	0	1	0	0
Harbour (%)	39	27	26	17	17	25	41	34	32
Domestic heating (%)	0	0	1	1	0	1	0	0	2

Stations	IS1	IS2	IS3	IS4	IR1	IR2	ITR	ITS	TU
PM10									
Total annual predicted concentration (µg m^{-3})	**27.8**	**28.6**	**5.1**	**4.9**	**7.7**	**5.4**	**5.2**	**4.7**	**9.4**
Industrial activities (%)	89	91	70	76	84	80	67	75	75
Road Transport (%)	4	4	19	10	10	9	22	11	13
Harbour (%)	7	4	8	4	4	7	10	12	8
Domestic heating (%)	1	1	3	10	2	4	2	2	4
PM2.5									
Total annual predicted concentration (µg m^{-3})	**20.9**	**21.9**	**4.1**	**3.8**	**5.9**	**4.3**	**4.0**	**3.7**	**7.5**
Industrial activities (%)	87	90	66	72	81	77	63	71	71
Road Transport (%)	4	5	21	11	12	10	23	12	14
Harbour (%)	8	5	9	5	5	8	12	15	10
Domestic heating (%)	1	1	4	12	3	5	2	2	5
C6H6									
Total annual predicted concentration (µg m^{-3})	**1.4**	**1.3**	**0.5**	**0.4**	**0.5**	**0.5**	**0.3**	**0.4**	**0.9**
Industrial activities (%)	76	77	34	40	55	57	56	46	40
Road Transport (%)	20	20	59	38	39	34	38	48	52
Harbour (%)	2	1	1	1	1	1	2	2	1
Domestic heating (%)	2	2	6	22	6	9	5	4	7

Industrial activities have been found to be the principal contributor to the total SO2 concentration regardless of stations. Harbour activities represented the second largest contribution.
Industrial activities represented the principal contributor to the total C6H6 concentrations for the stations closest to the industrialized area. In the other ones traffic and domestic heating were important contributing sources.

Industry and traffic emissions were, for the most part, responsible for NOx simulated concentrations, while primary PM10 and PM2.5 simulated concentrations appeared to be mainly linked to industrial emissions.

In order to evaluate the accuracy of the system used in this study, the model predictions of NOx, SO2, PM10, PM2.5 and C6H6 were compared with measurements taken in the air quality stations. In particular, the modelling performances were evaluated using some statistical indexes: mean, fractional bias (FB), root mean square error (RMSE), normalised mean square error (NMSE), correlation coefficient (CC) and the fraction of predicted values within a factor of 2 of observed values (FA2). When comparing observed data with model results, it must be recognised that, 1) the measurements are taken at discrete locations, while the calculated values are representative for a volume grid cell of 500 × 500 x 20 m, and 2) the chemical reactions are not included.

Figure 3 shows, as an example, the scatter plots measured vs. predicted concentrations for the yearly mean value of NOx and PM10, respectively. The dotted gray lines in the figures denote the accuracy range (30% for NOx and 50% for PM10) established by the Italian regulation (D.lgs n.155, 2010), for the modelling of the concentration. These lines represent the legal limits for the results as regard the yearly mean value.
The scatter plots showed that for NOx there is a slight tendency of the model to underestimate the mean concentration values, especially for the traffic TU, probably due to the incorrect temporal and spatial disaggregation of traffic flows over the simulation grid. For PM10 the underestimation, more considerable in particular at the monitoring stations located at a greater distance from industrial areas, is likely due to the regional background contribution, which was not taken into account by Lagrangian models.

(a) (b)

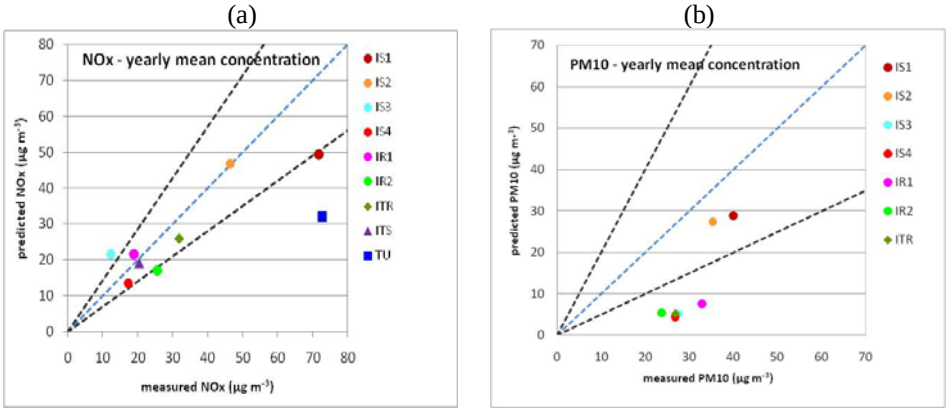

Figure 3. Scatter plots of measured vs predicted yearly mean concentrations of (a) NOx and (b) PM10.

Statistical indexes for NOx are shown in Table 4. Correlation is quite good especially for the IS1 and IS2 stations close to the industrial site. The worst results were obtained at stations far from the main sources (industry), maybe due to an incomplete representation of local emission sources not related to industry.

Table 4. Statistical indexes for NOx

Stations	IS1	IS2	IS3	IS4	IR1	IR2	ITR	ITS	TU
NOx									
Observed mean (μg m^{-3})	71.8	46.4	12.4	17.3	18.9	25.7	31.9	20.5	72.7
Predicted mean (μg m^{-3})	49.4	46.8	21.6	13.5	21.6	17.0	25.9	19.0	32.0
FB	-0.37	0.01	0.54	-0.25	0.14	-0.41	-0.21	-0.07	-0.78
RMSE	69.15	53.80	29.42	24.23	38.61	32.01	34.23	33.13	74.78
NMSE	1.35	1.33	3.23	2.51	3.65	2.35	1.42	2.82	2.40
CC	0.32	0.34	0.29	0.12	0.16	0.28	0.23	0.10	0.26
FA2	0.50	0.56	0.51	0.31	0.47	0.25	0.46	0.31	0.23

ACKNOWLEDGMENTS

The authors acknowledge ENEA for providing meteorological data produced within the MINNI Project over the south Italy domain and for the year 2007.

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