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MODELING THE LEVEL OF AIR POLLUTION USING EMISSION INVENTORIES

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ABSTRACT

This paper presents an approach for atmospheric pollution monitoring modeling based on the anthropogenic emissions. We focus on the analysis of NO_2 *emissions from the urban area of Braila city during the period 2009-2013. In order to predict the yearly spatial distribution of* NO_2 *emission we created five maps using data from the Environmental Protection Agency Braila by METI-LIS dispersion model. We observed that NO2 emissions decrease from 4.078 t in 2009 to 1.191 t in 2013.*

Keywords: NO_2, atmospheric pollution, emission inventories, dispersion model

1. INTRODUCTION

The NO_2 is considered an elementary pollutant in preparing the appraisal of the air quality index, and its excessive presence may determine important environmental and health problems. Whereas the contribution of NOx as a precursor of a numerous toxic pollutants and the repercussions that short - and long-term exposure to NO_2 pollution can cause to public health, it is obvious that maintaining NO_2 concentrations at reduced levels will ensure important advantages for the human health [1].

Most of NO_2 emissions from the atmosphere come from auto vehicles internal engine combustion exhaust plus emissions from the industry and heating power plants that use fossil fuel [2]. Significant increases of traffic levels in urban zones were observed, thus concerns regarding the concentrations of transportation have generated the necessity for a consistent regulatory scheme for managing air quality especially from urban areas [3].

The decrease of nitrogen oxide ($NOx=NO+NO_2$) emissions has been one of the significant objectives for improving air quality in Europe. The EU legislation regarding air pollution was improved by including the latest scientific and technological developments, yielding the publication of the Directive 2008/50/EC on ambient air quality and cleaner air for Europe. The EU First Daughter Directive (99/30/EC) established a yearly mean limit of

40 μg/m^3 and an hourly limit of 200 μg/m^3 that must not be exceeded more than 18 times a year [4].

Nitrogen dioxide is currently a problem for numerous cities due to its noxiousness and crucial role in the formation of tropospheric ozone [5, 6]. Mainly, in combustion processes, basically in engines of motor vehicles, NO_2 and NO are mostly formed and emitted immediately [7].

The directly emitted NO (NO direct), is transformed in the atmosphere, by O_3 or peroxy (RO_2) radicals into NO_2, which is called further NO_2 (indirect):

$$NO+O_3; (RO_2) \rightarrow NO_2 + O_2; (RO) \quad (1)$$

In the presence of sunlight, NO_2 is partially photolysed back to NO:

$$NO_2 \text{ (sunlight } (O_2)) \rightarrow NO+O_3 \quad (2)$$

Based on the assumption that the RO_2 photochemistry is insignificant and that the background O_3 concentration is uniform, the level of oxidants (OX) is given by [8]:

$$NO_2 + O_3 = OX \quad (3)$$

Dispersion models estimate the circulation of pollutants in air to simulate the spatial distribution of emission concentrations and are widely used to calculate the spatial distribution of a pollutant concentration. They have been classified according to the temporal and spatial scale, i. e. macroscale (measured in thousands of kilometres and days), mesoscale (measured in tens-hundreds and hours), and microscale (less than 1 km and minutes) [9]. Models also can describe near-field effects (<1 km), short-range effects (<10 km), intermediate transport (10-100 km), long-range effects (>100 km), and global effects [22]. Dispersion models have been categorized as statistical, deterministic, mathematical and physical modeling [11]. Even if urban traffic induces turbulence and could be an important factor in pollutant dispersion, these are not taken into consideration. Wind direction, wind speed and atmospheric stability have, on the other hand, a major contribution on dispersion. If meteorological conditions are not favorable, NO_2 concentration might increase even when NOx source emissions decrease [12].

According to emission calculations inventory of the Environmental Agency from Braila, the decrease in total NOx emissions between 2008 and 2013 is approximately 60% [13]. In Braila, Romania, nitrogen oxide emissions are still principally generated by road traffic [14]. This paper is focused on presenting a method to define the spatial and temporal variation of the NO_2 emission levels using a dispersion model.

2. DATA AND METHOD

The procedure used for calculation and reporting emissions is in conformity with the Guide established in 2013 by the Executive Committee of The Convention on Long-Range Transboundary Air Pollution. Total national emissions are predicted according to the EMEP-CORINAIR (COoRdination d'INformation Environmentale) guide - book, the IPCC guidelines and the Good Practice Guidance and classified accordingly to SNAP (Selected Nomenclature for Air Pollution) [15-17].

CORINAIR is the most significant European air emission inventory programme using a precise emission calculation procedure and software for collecting data and processing. In CORINAIR inventory 28 distinctive emission species are predicted in 11 principal sectors and subsequent divided in further detailed intermediate and third levels. The emission sources are considered as large point sources in conformity with appropriate CORINAIR definitions.

In CORINAIR, the traffic emission computing is divided in two main groups: Road traffic (SNAP group 07) and Other sources and machinery (SNAP group 08). As an additional part of CORINAIR, The COPERT (Computer Programme to calculate the

Emissions from Road Transport), a computer software has been created to estimate the road traffic emissions [18-19].

The calculation algorithms of the vehicles' emissions contain a combination of firmed technical data (e.g. emissions factors) and activity data (e.g. number of vehicles per category, per unit time) [20].

The emissions of the traffic category rely on a multitude of factors such as the distance that each vehicle circulate, its speed (or road type), its age, engine size, engine's technology (catalytic, non-catalytic vehicles, open loop, uncontrolled vehicles) and weight.

The basic formula for the estimation of emissions is the following:

Emissions [g] =emission factor [g/km]·vehicle kilometers per year [km] (4)

The dispersion of NO_2 was calculated using the METI–LIS program, software developed originally by the Japan Ministry of Economy, Trade and Industry. The METI-LIS 2.03, is a Gaussian dispersion model which calculates the manner how pollutant concentrations are distributes by cause of dispersion. Dispersion conditions are influenced by meteorological factors and by volume of emitted pollutant. Wind speed and direction, solar radiation and atmospheric stability have a significant influence on pollutant dispersion [21]. The METI-LIS soft uses a Gaussian plume equation that considers steady-state conditions. It can be applied to point or line sources. The most frequently used technique to integrate the point – source plume equation is Simpson formula of computing numerical integrals.

The NO_2 data-set from the Local Environmental Protection Agency was prepared to match the pattern appropriate by the model. Wind direction (degrees from N, clockwise) and wind speed, temperature and solar radiation were the meteorological factors required for our study. Sources with line-shaped characteristics are computed by numerically integrating the equation, (5)

$$C(x,y,z)=\frac{Q}{2\pi\sigma_y u}\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left[\exp\left(-\frac{(z-He)^2}{2\sigma_z^2}\right)+\exp\left(-\frac{(z+He)^2}{2\sigma_z^2}\right)\right] \quad (5)$$

where C is the concentration (g/m³), x is the downwind distance from the emission source (m), y is the crosswind distance from the emission plume centerline (m), z is the distance above the ground level (m), Q is the pollutant emission rate (g/s), He is effective plume-rise height, u is wind speed (m/s), σ_y is horizontal dispersion width (P-G curve) (m), σ_z is vertical dispersion width (P-G curve)(m).

3. RESULTS AND DISCUSSION

Local inventories are collected by the Local Environmental Protection Agency and centralized into the national database, depending on each emission source by sector. Every sector consists of individual categories (e.g. transport) and subcategories (e.g. passenger cars, light/heavy duty vehicles, two wheelers). Table 1 presents the NO_2 annual average data for each SNAP sector for period 2009-2013.

As reported by the local emission inventory, road transport, combustion in energy and transformation industries and non-industrial combustion plants are the most important contributors to air pollution, in Braila.

Zero NO_2 emissions can be observed as concerns Group 5, Extraction and distribution of fossil fuels and geothermal energy, Group 9, Waste treatment and disposal, Group 10 Agriculture because these activities are very restricted or inexistent in studied area.

Table 1 NO_2 local emissions inventory, 2009 – 2013, according to SNAP Group [tonnes]

Group	CORINAIR sector	2009	2010	2011	2012	2013
01	Combustion in energy and transformation industries	895.02	395.94	572.10	172.86	128.02
02	Non-industrial combustion plants	115.88	169.57	207.44	178.00	100.98
03	Combustion in manufacturing industry	33.02	83.59	63.72	13.36	14.56
04	Production processes	0.04	0.28	0.25	0.12	0.08
05	Extraction & distribution of fossil fuels and geothermal energy	0.00	0.00	0.00	0.00	0.00
06	Solvent and other product use	20.47	10.14	27.49	12.45	10.12
07	Road transport	3000.17	3240.12	3609.95	1910.43	1124.19
08	Other mobile sources and machinery	34.50	93.79	58.68	81.28	23.46
09	Waste treatment and disposal	0.00	0.00	0.00	0.00	0.00
10	Agriculture	0.00	0.00	0.00	0.00	0.00
11	Other sources and sinks	0.00	0.00	0.00	0.00	0.00

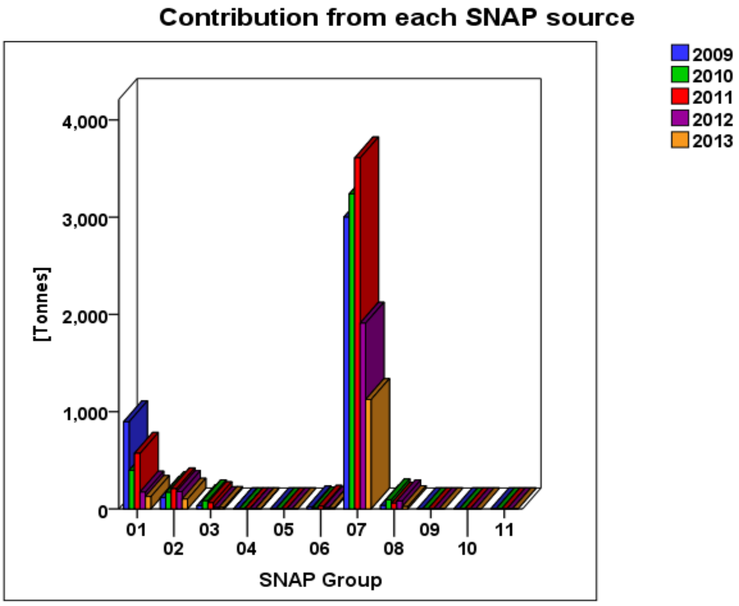

Fig. 1. NO_2 annual contribution of anthropogenic emissions by type of activity

Figure 1 presents the NO_2 annual contribution of anthropogenic emissions by type of activity, from 2009 to 2013. The NO_2 emissions were stated using the declarations supplied by operators inventoried.

The analysis of the evolution of emissions from anthropic activities, determined according to SNAP source, shows that emissions have increased until 2011 and constantly have decreased afterwards, until 2013. This is seen for all 11 types of activity and results from reducing the activity of important industrial and economic operators. The production of Energy sector was reduced from 2012 due to a significant decrease in activity of the two large combustion installations (respectively Combined Heat and Power Plant Brăila and Thermoelectric Electrocentrale Brăila. The largest amount of pollutants in the atmosphere, from 2009 to 2013, result from road traffic (78.56% of total emissions, i.e. 12884.87 tons) and only 13.19%, namely, 2163 tons come from combustion in energy and transformation industries.

As stated in local inventory the principal contribution to air quality has the SNAP Group 7, Road transport. Even if the period is very short, a downward trend is observed, from 3000.17 t in 2009 to 1124.19 t in 2013. A significant decrease is observed in 2012, when the amount of NO_2 was 1910.43 t, generally less than 52% of the NO_2 estimated in 2011. Fig. 2 presents the NO_2 emission trend in Braila, from 2009 to 2013.

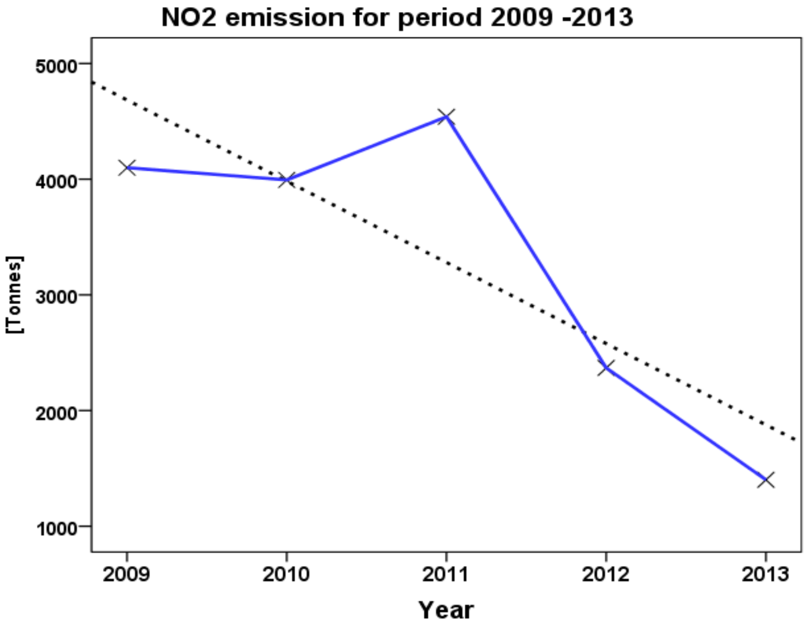

Fig. 2 NO_2 emission trend in Braila, from 2009 to 2013

The largest NO_2 emission can be observed in 2011, i.e. 3609.95 t (the first year of the analysis), while in 2013 the NO_2 was just 1124.19 t. The decreasing trend of NO_2 is explained by the industrial and economic crisis and likewise it is a result of increasing number of vehicles equipped with oxidation catalysts.

In order to obtain a clearly image of the spatial dispersion of NO_2 five maps containing the yearly averages of NO_2 emission from the urban area of Braila city were plotted.

The METI-LIS soft was used to model the dispersion of a NO_2 emission using a 50 × $50m^2$ spatial grid in urban area of Braila. Yearly dispersion maps obtained using METILS are presented in Fig. 3. The highest NO_2 emission can be observed throughout the main roads and in their nearly surroundings, evidently related to the traffic. According to the local emission inventory, the NO_2 quantity progressively decreases. High NO_2 is observed throughout all main streets in 2009, with some frequency close to the suburb of the city. These spots are also seen in 2010, 2011, 2012 and 2013 and appear, progressively, in the city center.

The METI LIS dispersion model indicates a higher NO_2 values in the vicinity of the main streets and some abrupt changes. One explanation is that the spatial distribution of emissions depends strongly on the meteorological data, i.e. on wind speed and direction.

Likewise, a careful visual examination to the maps indicates that spots of NO_2 arise for the most part near cross-roads and, presumably, near traffic lights.

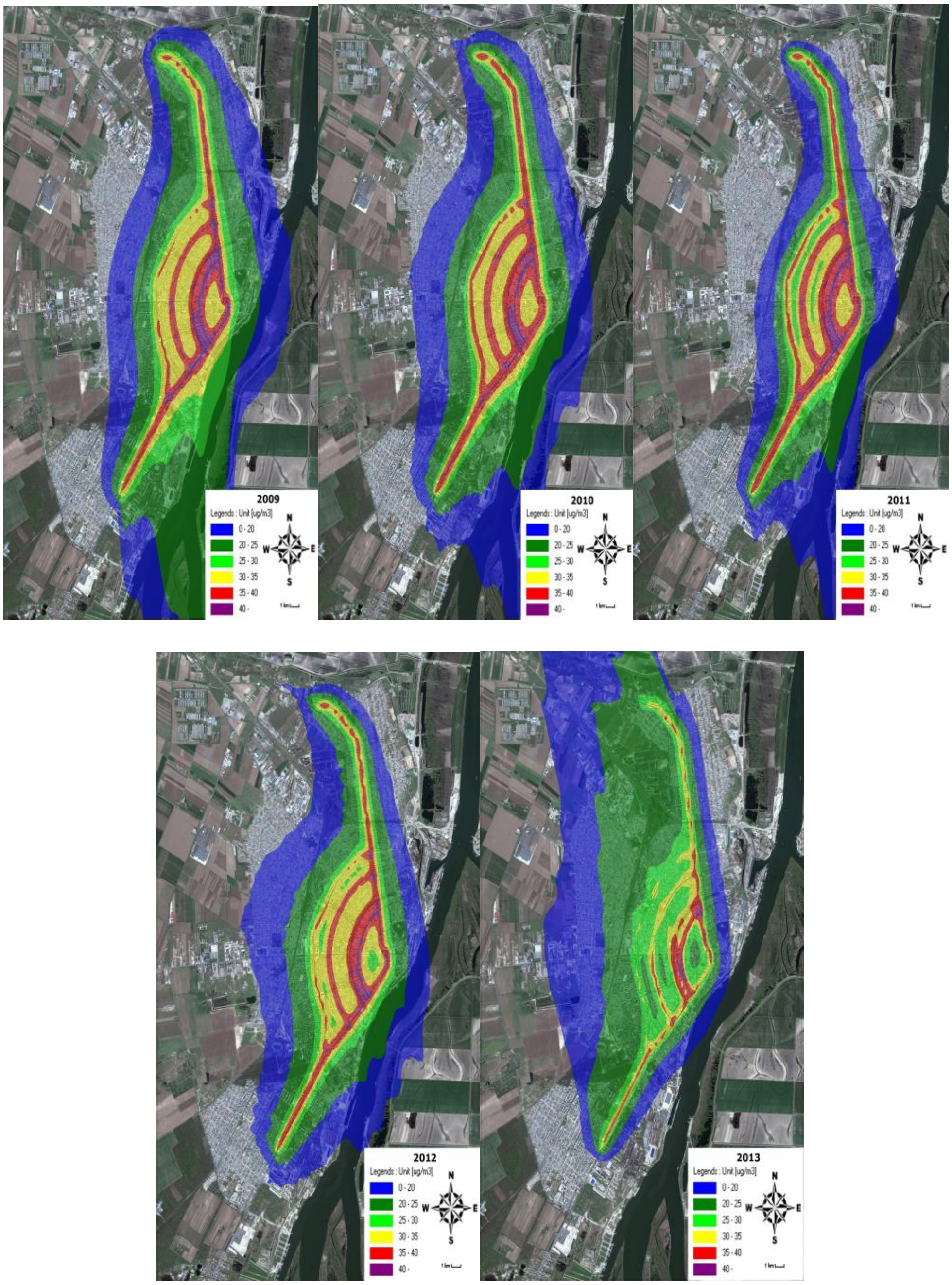

Fig. 3. Dispersion of NO_2 for 2009-2013. NO_2 emission between 0 and 20 µg/m^3 are shown in blue color, 20-25 µg/m^3 in dark green, from 25 to 30 µg/m^3 in light green, 30-35 µg/m^3 in yellow, 35-40 µg/m^3 in red and over 40 µg/m^3, in violet. Note that the intense green to the SE of the map is due to the superposition with the dark colors used for the Danube.

The decrease of NO_2 quantity in 2012 and 2013 may be the result of a significant decrease of the polluted area, which is wide, for instance in 2010 and 2011, but small in 2012 and 2013. This is a consequence of meteorological variation, in addition to the decreasing number of anthropic sources. Very likely, a SW wind carries NO_2 loading to the southern part of the city in 2009-2013. A substantial reduction of NO_2 is noticed basically at the peripheries of the urban area, to the West. Roads that were heavily polluted in 2011 tend to be increasingly less affected by traffic.

4. CONCLUSIONS

The Corinair inventory contains the most consecrated set of emission estimation techniques used in air pollution researches in the geographical area of Europe.

NO_2 emissions and yearly averages of meteorological factors were used to create five maps of the NO_2 dispersion in an urban area of Braila. We used the METI-LIS soft for a period of five years, 2009-2013. As expected, METI-LIS maps indicate that the highest NO_2 value refers to linear sources connected with traffic.

A significant diminution of yearly averages of NO_2 is observed from 2012 to 2013. NO_2 spots also abide and are associated with semaphores or crossroads. The dispersion area of NO_2 is also influenced, principally, by meteorological parameters but also by increasing number of car engines equipped with particle filter.

A local and national inventory helps to clarify the environmental priorities, develop the air quality modeling and establishes the efficacy of policy actions as respects the protection of human health and the environment.

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REFERENCES

[1] Mavroidis, I., Chaloulakou, A, 2011, Long-term trends of primary and secondary NO2 production in the Athens area. Variation of the NO2/NOx ratio. *Atmospheric Environment*, 45, pp. 6872–6879

[2] Collett, R.S., Oduyemi, K., Davidson, B., 1997, An investigation into ambient levels of traffic related pollutants in urban centres: a case study of the Seagate, Dundee, United Kingdom, *Journal of Environmental Management*, 51, pp. 289-304.

[3] Grice, S., Stedman, J., Kent, A., Hobson, M., Norris, J., Abbott, J., Cooke, S., 2009, Recent trends and projections of primary NO2 emissions in Europe, *Atmospheric Environment*, 43 (13), pp. 2154-2167.

[4] ***EC (European Commission), 2008, Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 On ambient air quality and cleaner air for Europe, *Official Journal of the European Union*, L 152/1.

[5] Seinfeld J.H, Pandis S.N, 1998, Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, New York: John Wiley & Sons.

[6] Kurtenbach, R., Kleffmann, J., Niedojadlo, A., Wiesen, P., 2012, Primary NO2 emissions and their impact on air quality in traffic environments in Germany, *Environ. Sci. Eur.*, pp. 21-24.

[7] Clapp L.J, Jenkin M.E., 2001, Analysis of the relationship between ambient levels of O3, NO_2 and NO as a function of NOx in UK, *Atmospheric Environment*, 35, pp. 6391–6405.

[8] Suzuki, H., Miyao, Y., Nakayama, T., Pearce, J.K., Matsumi, Y., Takahashi, K., Kita, K., Tonokura, K., 2011, Comparison of laser-induced fluorescence and chemiluminescence measurements of NO_2 at an urban site, *Atmospheric Environment*, 45, pp. 6233-6240.

[9] Khare, M., Sharma, P., 2002, Modelling Urban Vehicle Emissions, WIT Press.

[10] Carbonell, L.T., Mastrapa, G.C., Rodriguez, Y.F., Escudero, L.A., Gacita, M.S., Morlot, A.B., Montejo, I.B., Ruiz, E.M., Rivas**,** S.P., 2013, Assessment of the Weather Research and Forecasting model implementation in Cuba addressed to diagnostic air quality modeling, *Atmospheric Pollution Research*, 4, pp. 64-74.

[11] Kastner-Klein, P., Fedorovich, E., Ketzel, M., Berkowicz, R., Britter, R., 2003, The modelling of turbulence from traffic in urban dispersion models - Part II: evaluation against laboratory and full scale concentration measurements in street canyons, *Environmental Fluid Mechanics*, 3 (2), pp. 145-172.

[12] Pleijel, H., Klingberg, J., Back, E., 2009, Characteristics of NO_2 pollution in the city of Gothenburg, south-west Sweden - Relation to NOx and O_3 levels, photochemistry and monitoring location, *Water Air Soil Pollut. Focus*, 9, pp. 15-25.

[13] Environmental Protection Agency Braila, Environment Report Braila County 2013, Braila, Romania.

[14] Dragomir, C. M., Constantin, D.E., Voiculescu, M., Georgescu, L. P., Merlaud, A., Van Roozendael, M., 2015, Modeling results of atmospheric dispersion of NO2 in an urban area using METI–LIS and comparison with coincident mobile DOAS measurements, *Atmospheric Pollution Research*, 6, pp. 503-510.

[15] EEA (European Environment Agency), EMEP/EEA air pollutant emission inventory guidebook 2013, Published: 29 Aug 2013, 26 pages, ISSN 1725-2237

[16] *** EEA (European Environment Agency), Technical report, No 12/2013 EMEP/EEA emission inventory guidebook 2013, Technical guidance to prepare national emission inventories, ISSN 1725-2237.

[17] *** EC (European Commission) 2006, Guidance Document for the Implementation of the European PRTR, European Commission, May 2006.

[18] *** IPCC (2006), 2006 IPCC Guidelines for National Greenhouse Gas Inventories, IPCC National Greenhouse Gas Inventories Programme, Global Environmental Strategies (IGES), Hayama, Japan.

[19] Rypdal, K., Stordal, F., Fuglestvedt, J.S. and Berntsen, T., 2005, Bottom-up vs. top-down methods in assessing compliance with the Kyoto Protocol, Climate Policy 5, pp. 393-405.

[20] Kouchi, A., Okabayashi, K., Okamoto, S., Yoshikado, H., Yamamoto, S., Kobayashi, K., Ono, N., Koizumi, M., 2004, Development of a low-rise industrial source dispersion model (METI-LIS model), *International Journal of Environment and Pollution*, 21, pp. 325-338.

[21] *** METI. 2006, Low Rise Industrial Source Dispersion Model METI-LIS Model Ver. 2.03, Operation Manual. Ministry of Economy, Trade and Industry.

[22] Zanetti, P., 1990, Air pollution modeling—Theories, computational methods, and available software. New York: Van Nostrand Reinhold