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THE APPLICATION OF CALMET/CALPUFF MODELS IN AIR QUALITY ASSESSMENT SYSTEM IN POLAND

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Abstract: The accession of Poland to the European Union involved the need of regional air quality assessment and brought radical change in requirements towards the software tools used for assessment purposes. According to Polish law, a zone is an agglomeration of over 250 000 inhabitants, or a poviat (second level of local government administration in Poland), or a group of poviats, and assessment should consider both global and regional inflow of pollutants as well as the impact of local emission sources and significant sources in a voivodeship. These requirements have imposed a model range of over 250 km. Following an analysis of different models operating all over the world, the CALPUFF model together with the CALMET meteorological processor was chosen to be implemented in air quality assessment systems in Polish zones. This paper presents the results of model calculations performed within the air quality assessment in Mazowieckie voivodeship as well as compares them with the measurements obtained at automatic air monitoring stations.

INTRODUCTION

The requirements of Framework Directive 96/62/EC [7] and daughter directives have been transposed to the Polish Environment Protection Law of 27 April 2001 [22]. This law has systematized all the efforts concerning the development and implementation of a complex air quality assessment system in voivodeships and gave them final shape. Every year the Head of Voivodeship Council carries out the assessment of air pollutant level in a given zone and subsequently classifies the areas into three categories: A, B or C. In consequence of categorizing an area to the C class it is necessary to develop an air protection program including all the pollutants exceeding the limit value.

The Environment Protection Law [22] transfers the regulations applied in the European Union into Polish law and this way increases the role of the society in public life, granting every citizen the right to have access to the information possessed by the authorities. Also the Aarhus Convention of 25 June 1998 [6] on access to information, public participation in decision-making and access to justice in environmental matters imposes certain duties on public authorities.

In order to accomplish the above-mentioned tasks, it is necessary to design a voivodeship air quality assessment system.

The extensive air quality assessment system in a voivodeship has to provide people and public authorities with appropriate information and give basis for decision-making, leading to significant financial and social effects. The system should therefore be "live", constantly developing.

The extensive voivodeship air quality assessment system comprises three main blocks [10]:

- a measuring system
- a spatial analysis system
- an information and decision-making system.

The aim of the system is to provide information used for:

- planning and performing actions aimed at improving air quality in the areas where the adopted quality criteria are not met,
- maintaining status quo of air quality in the areas where it is good.

The measuring system and the spatial analysis system provide information that is essential for determining air quality in a given area. Moreover, the spatial analysis block provides tools for permanent improvement of the whole air quality assessment system and for creating spatial pollutant distribution forecast. On the basis of reliable data acquired from those two blocks it is possible to prepare indispensable information for citizens and authorities, who in turn will be able to initiate cautionary and pollution control procedures when necessary.

Irrespective of measuring methods, applied information obtained in this way, although the most reliable if certain conditions are fulfilled, determines concentration value only at the measuring spot. This information can be extrapolated to a representative area of a measuring station, understood as the area where pollutant concentrations are connected in certain way with the measured value. However, in each area there are such zones (usually predominating), for which it cannot be inferred about pollutant distribution on the basis of performed measurements. The diversification of concentrations in these zones can be determined only based on model calculations, since they allow to obtain information concerning the spatial diversification of concentrations in an entire area with reference to diversified time intervals. The value of modeling effects depends on two basic elements: the right choice of a model and the quality of data entered into it.

MATERIALS AND METHODS

To perform calculations of air pollution spreading for the purposes of an air quality assessment system the CALPUFF model [24] was chosen, recommended by the US EPA (United States Environmental Protection Agency), together with the CALMET meteorological model [25].

CALPUFF is a new generation Gaussian puff model, which operates in Lagrangian system and considers the geophysical data (land use type elevation, surface parametres) as well as the temporal and spatial variation of meteorological conditions in three dimensions. It is a multilayer, non-stationary model designed for calculating concentrations of many substances, which can determine the influence of emissions from different types of sources as well as of meteorological fields (temporally and spatially changeable) on transport, chemical transformation and pollution deposition. A plume of pollution emitted from a source is moved by puffs. In puff models [20] it is assumed that within each Δt time range pollution of mass equal to $\Delta M = E \cdot \Delta t$, where E stands for temporally changeable pollution emission intensity [20], is discharged into the atmosphere. At the same time the wind structure assumed does not have a form of a continuous air stream but is a sum of "puffs". A pollution puff of ΔM mass moves according to local, temporally changeable wind velocity vector and the pollution cloud diffusion is described by standard deviations of a Gaussian concentration distribution.

The CALPUFF model has found wide applications in many countries, both in air quality assessment systems [4, 18, 19], as well as in the assessment of the effect of industrial plants on the environment. In the analysis of air pollution spreading from existing and designed power plants the CALMET/CALPUFF models have been applied in the USA [1, 16, 21], Canada [8], China [11], Mexico [17], Turkey [12] and Italy [19]. The model has also been successfully used in Poland in regional air quality assessment systems [9, 10], air protection programs [29] and in the assessment of large emitters influence [21, 28].

In the case of calculations for the purposes of an air quality assessment system in an area as large as a voivodeship, the most essential model characteristics are:

- varied elevation and land use classes,
- varied meteorological conditions in a given area,
- temporally changeable point source, surface and traffic emissions,
- chemical transformations, dry and wet deposition,
- the ability to determine organic and inorganic aerosols taking into account the variability of ozone and ammonium background,
- boundary and initial conditions which is especially important in the case of $PM_{2.5}$ and aerosols,
- spatial range of model operation up to 300 km,
- temporal range of the model: from 1 hour to 1 year,
- possibility to determine the share of single sources or their groups, or types in spatial distributions of pollution concentrations, first of all in the areas where limit values are exceeded,
- constant development of the system,
- access (preferably free) to the program sources and documentation.

The basic sources of data for the CALMET meteorological model are measuring results from synoptic, precipitation, and aerologic stations. However, on account of a relatively poor measuring network, first of all in the vertical layer of the atmosphere, but also in the ground layer, as well as in the precipitation measurement, solutions based on meteorological fields determined by larger scale models are preferred. As an example we might take the calculations that were made for the purposes of air quality assessment in Mazowieckie voivodeship for the year 2008.

A following calculation scheme that was applied: WRF → CALMET → CALPUFF was based on the model nesting conception. The WRF model [30] is a mesoscale numerical meteorological model, a dynamic model with data assimilation, designed for simulating and forecasting atmospheric circulation. As the input, the data information coming from the American NCEP/NCAR Reanalysis project are used. These data consider all the measuring information from ground, aerologic and precipitation measuring networks, as well as from surveys and satellite observations.

The WRF model is widely applied [15], among others, in the USA, Europe, Canada and Australia as a source of meteorological data for air quality assessment systems.

The analysis of the application of the CALPUFF model for the purposes of air quality assessment is presented below on an example of model calculations for Mazowieckie voivodeship in 2008. Figure 1 presents the chosen modeling area – Mazowieckie voivodeship. The picture also shows the location of automatic air monitoring stations. The results obtained on those stations will be used in the verification of the model.

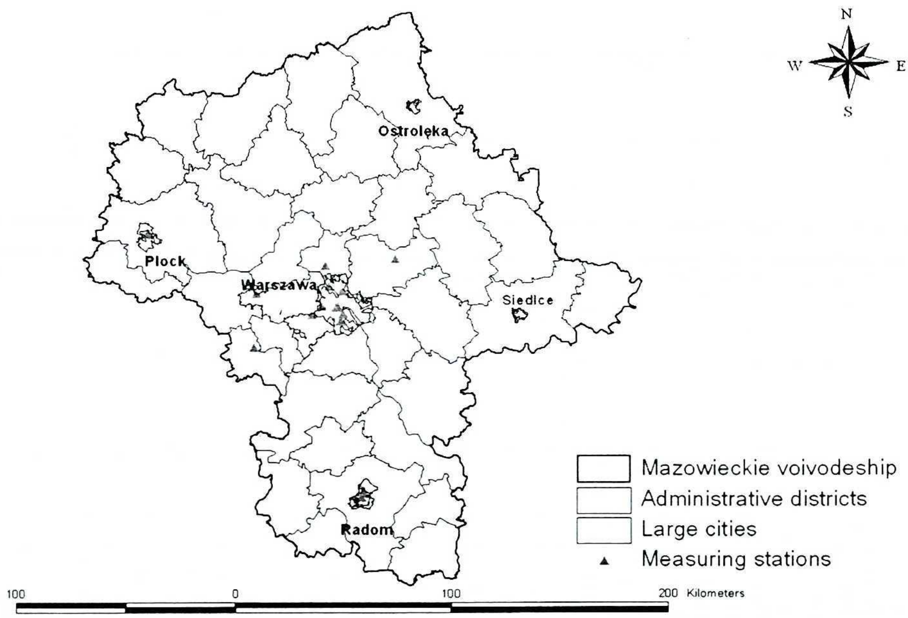

Fig. 1. Research area

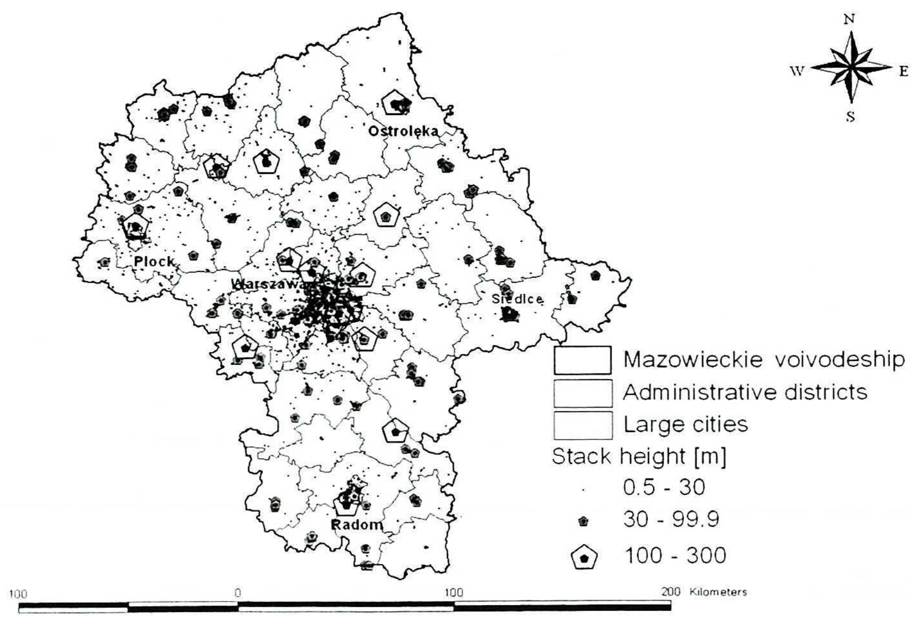

Fig. 2. Point sources in Mazowieckie voivodeship in relation to stack height

For the chosen area a detailed emission source inventory was performed. 4239 industrial, energetic and technological sources were inventoried, as well as 8049 municipal emission sources, 9872 traffic emitters and 333 sources of agricultural emission. The location of point sources in relation to stack height is presented in Figure 2.

Figures 3, 4 and 5 show examples of surface emission distributions (Fig. 3 – PM_{10}), linear emission distributions (Fig. 4 – NO_x) and agricultural emission distributions (Fig. 5 – NH_3).

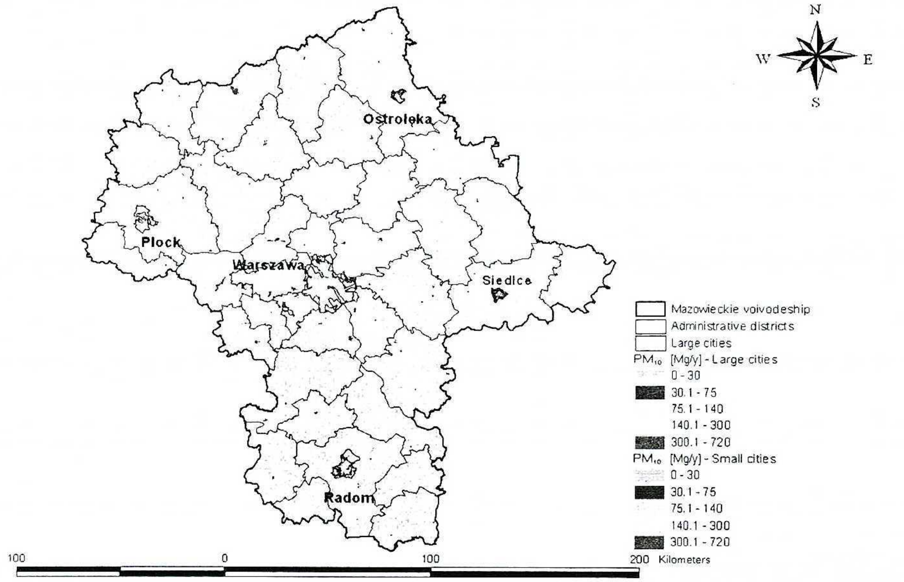

Fig. 3. Surface emission cadastre of PM_{10} [Mg/y] in Mazowieckie voivodeship divided into large cities and the rest

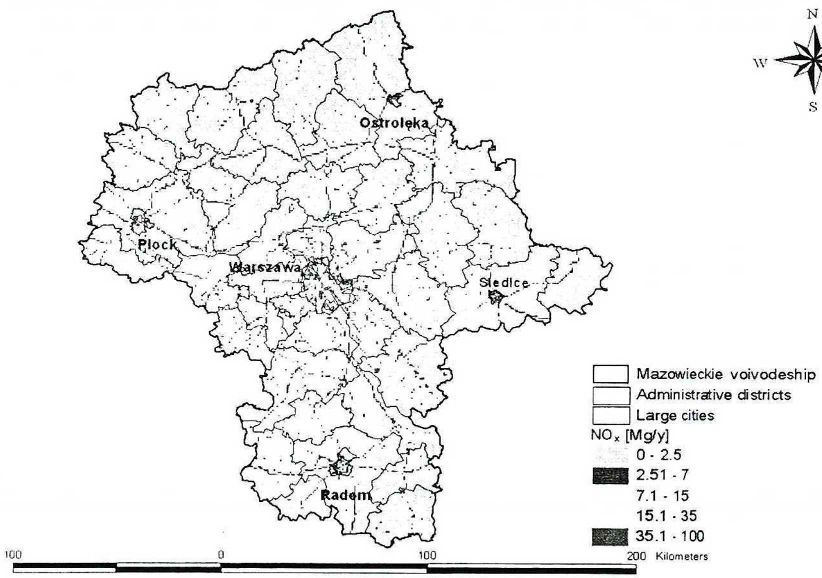

Fig. 4. Linear emission cadastre of NO_x [Mg/y] in Mazowieckie voivodeship

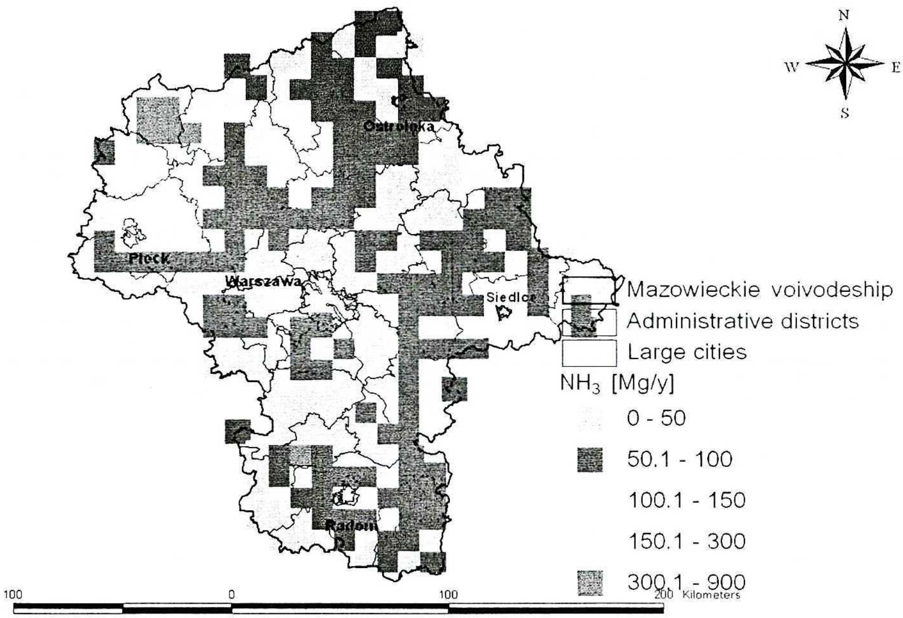

Fig. 5. Agricultural emission cadastre of NH_3 [Mg/y]

For all the types of emitters time variability was determined, characteristic for a given type of emission – seasonal and daily, for point, surface and linear sources, and, additionally, for days of the week for traffic sources. Furthermore, the fact that emissions depend on temperature was taken into account. Point sources were entered into the model in the form of individual sources with specific technical parameters and emission rate, traffic and municipal sources – in the form of a cadastre in an area of 1 km x 1 km and agricultural sources – in the form of a cadastre in an area of 10 km x 10 km. Additionally for Warsaw the cadastre was compacted to 250 m x 250 m for traffic sources and 500 m x 500 m for municipal sources, and in the rest of the larger cities the surface cadastre for traffic emission was 500 m x 500 m.

Particularly great attention was paid to determining emissions from traffic sources, especially PM_{10} and $PM_{2.5}$ emissions. Engine emissions were considered, as well as friction emissions (tyres, road surface, brake pads) and emissions from dust (resuspended during motor-vehicle traffic).

The CALPUFF model configuration included chemical transformations such as ozone changeability (based on automatic measurements) and ammonium background, as well as dry and wet deposition. This is a basic condition for correct determination of nitrogen and sulphur oxides as well as PM_{10} and $PM_{2.5}$.

Pollution inflow from outside the area of calculation was considered by adding boundary concentrations to the CALPUFF model, thanks to what temporal and spatial variability was introduced. Boundary conditions for all the substances, primary and secondary (nitrates and sulphates), as well as ammonium, were determined following the procedure described in the paper [5], according to which in the external cells of a meteorological grid annual mean values of substances are presented together with their monthly variability. Due to the lack of reliable measurement data, first of all from the eastern and southern parts of the modeling area, the results of the EMEP model [26, 31] were used for determining the value in the external cell.

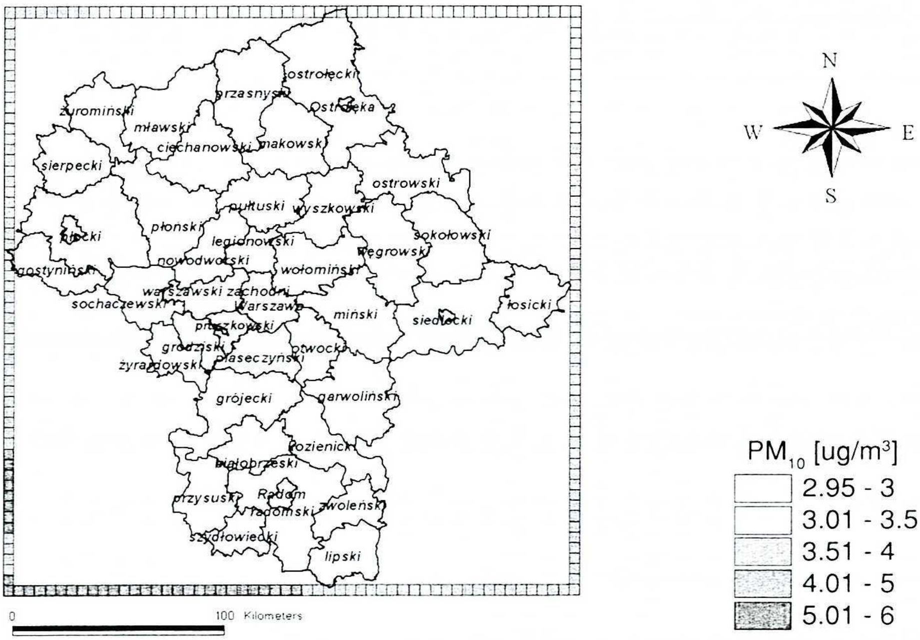

Fig. 6. Boundary conditions for PM_{10} – annual mean value

RESULTS

Within the modeling for the purposes of air quality assessment in Mazowieckie voivodeship, concentration distributions of 7 pollutants were determined: SO_2, NO_2, NO_x, CO, PM_{10}, Pb and C_6H_6. In this paper we will present modeling results for SO_2, NO_2, and PM_{10}. The calculations were carried out for the year 2008.

In the assessment of the quality of the results obtained we applied dispersion charts of calculated values versus measured values, compared time series of calculated values and measured values and applied a statistical package proposed by Juda-Rezler [13, 14]:

1. NMB – Normalized Mean Bias,
2. NMSE – Normalized Mean Squared Error,
3. RMSE – Root Mean Squared Error,
4. RMSEs – Systematic part of RMSE,
5. RMSEu – Unsystematic part of RMSE,
6. r – correlation coefficient,
7. IA – index of agreement; an alternative to correlation coefficient, more resistant to divergent values,
8. FB – Fractional Bias,

9. EXV – Explained Variance – variance explained by the model; it is a measure determining what part of measurement variance is explained by the model; for good models EXV ≥ 0.3.

The requirements that should be met by air quality modeling results are stipulated in the Order of the Minister of the Environment of 17 December 2008 on the assessment of the level of substances in the air [23]. These requirements are presented in Table 1.

Table 1. Required modeling accuracy

Accuracy	SO_2, NO_2, NO_x	Suspended dust, PM_{10} and Pb	Benzene	CO	Ozone
1 h concentration mean	50–60%		–	–	50% by day
8 h concentration mean	–	–	–	50%	50%
24 h concentration mean	50%	–	–	–	–
Annual concentration mean	30%	50%	50%	–	–

Figure 7 shows the results of modeling performed with the WRF-CALMET-CALPUFF model system for SO_2. The figure presents the distribution of the daily mean concentration 4[th] maximum (percentile 98.9), with limit value amounting to 125 μg/m^3.

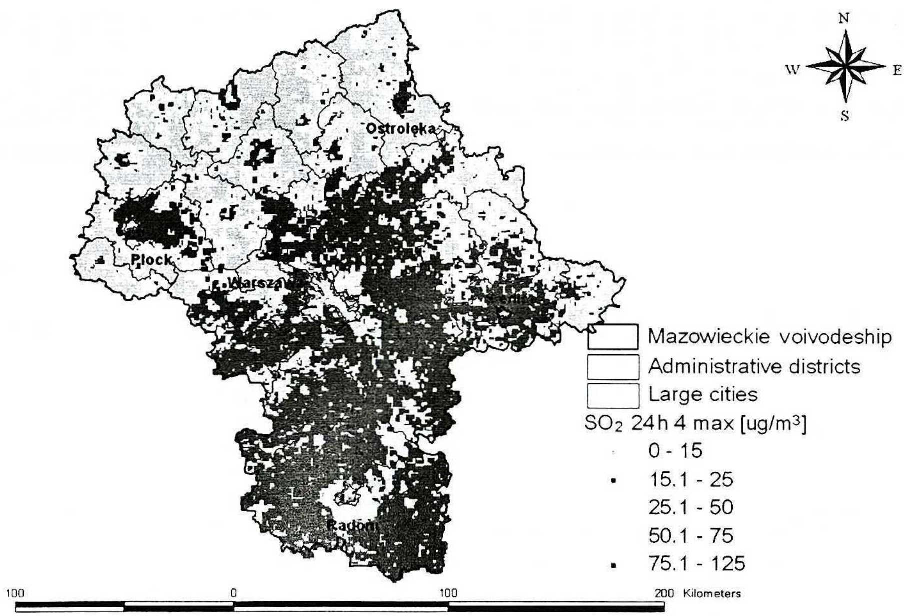

Fig. 7. SO_2 daily mean concentrations in [μg/m^3], percentile 98.9, in Mazowieckie voivodeship in 2008

As can be seen from Figure 7, SO_2 concentrations for the majority of Mazowieckie voivodeship are characterized by very low values (0–15 μg/m^3) in the north of the area and moderate values (15–50 μg/m^3) in its central-southern part. Higher concentrations appear only in cities (75–120.6 μg/m^3) although the values still do not exceed the upper threshold for estimated SO_2. Limit value was not exceeded.

The scatter plot of the predicted and observed values for percentile 98.9 of daily mean SO_2 concentrations with the values of statistical measures that were applied are presented in Figure 8.

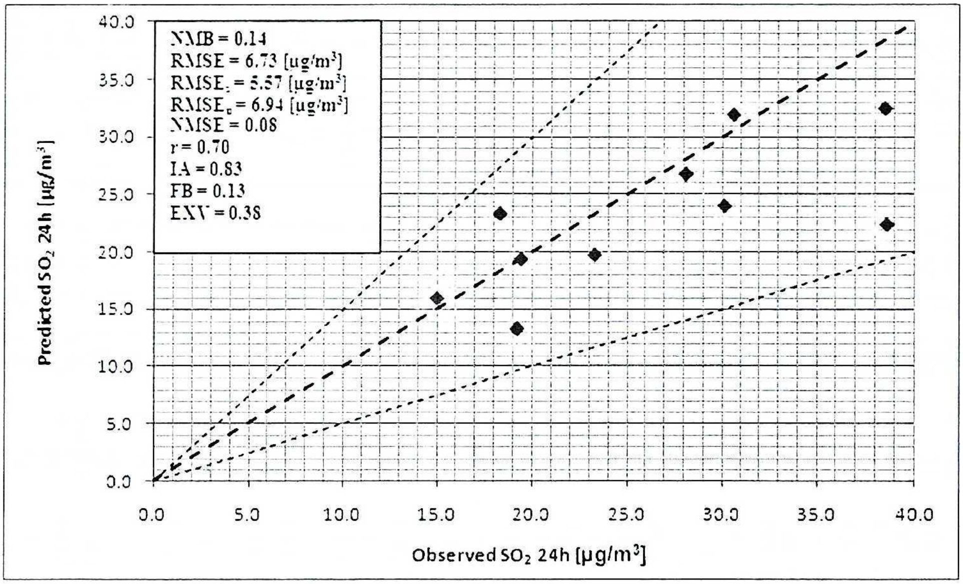

Fig. 8. Predicted and observed values for percentile 98.9 of daily mean SO_2 concentrations in [µg/m³] in Mazowieckie voivodeship; dotted lines show the variability scope imposed by the Minister's order (± 50%) and ideal conformity of the model with the measurements (the middle line)

The comparison of predicted SO_2 daily mean concentration time series with the values measured at the station in Piastów is presented in Figure 9.

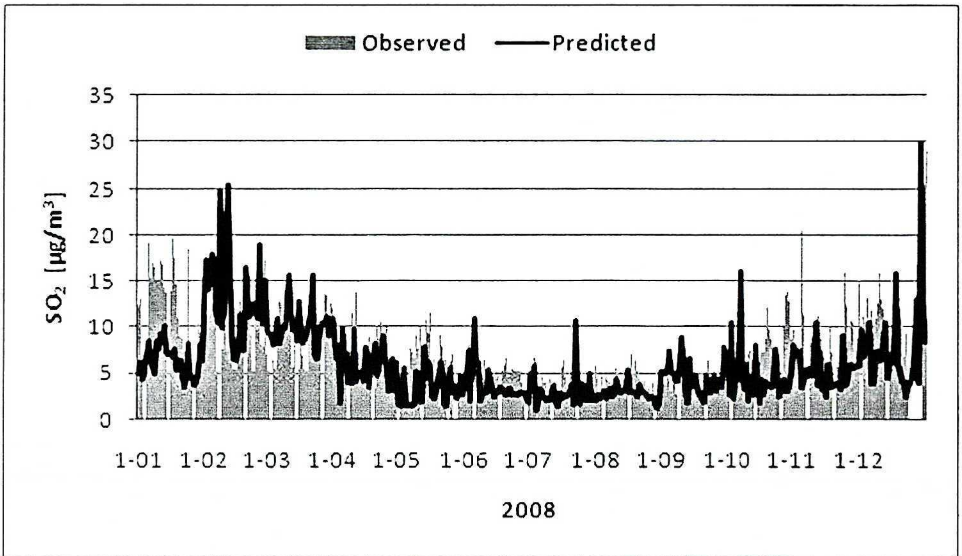

Fig. 9. Daily means of SO_2 concentrations, estimated and observed, in [µg/m³] time series in Piastów, 2008

The obtained statistical values assessing the model performance for SO_2 (shown in Fig. 8) demonstrate that the CALPUFF model determines SO_2 characteristics in a very satisfactory way. Both the high correlation coefficient (r = 0.7), as well as the index of agreement (IA = 0.83) indicate a strong relation between model values and measured values. All the modeling results stay within the acceptable range of modeling results variability relative to measuring results required in the order of the Minister of the Environment. Very small values of normalized and fractional bias as well as normalized mean square error were recorded: NMB = 0.14, FB = 0.13, NMSE = 0.08, but a high value of explained variance was obtained: EXV = 0.38. The concentration time series presented in Figure 9 indicate that both in terms of value, as well as in terms of time, the model keeps up with observed values. Therefore, all the results presented allow us to accept that the model performs very well in the case of SO_2.

Figure 10 presents the modeling results from the WRF-CALMET-CALPUFF system of models for NO_2. The picture shows the distribution of the 19^{th} hourly concentration maximum (percentile 99.8), with limit value amounting to 200 μg/m³.

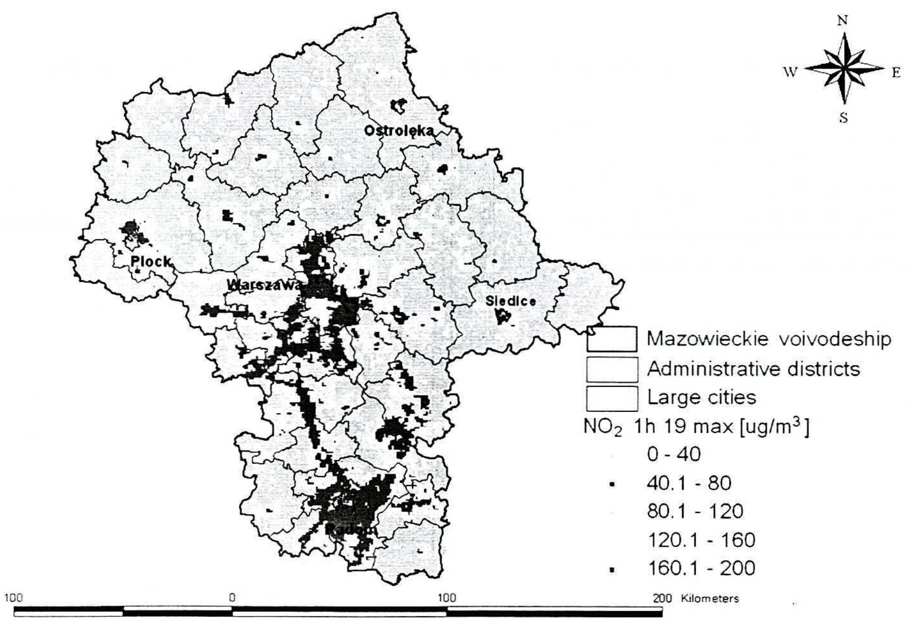

Fig. 10. NO_2 daily mean concentrations in [μg/m³], percentile 99.8, in Mazowieckie voivodeship, 2008

As seen from Figure 10, NO_2 concentration values are very low (0–40 μg/m³) in the majority of Mazowieckie voivodeship area and moderate (40.1–80 μg/m³) in the area around larger cities (Warsaw Agglomeration, Radom). Higher concentrations occur only in cities (80.1–160 μg/m³), however they still do not exceed the upper threshold for estimated NO_2. The limit value was not exceeded anywhere. The scatter plot of the predicted and observed values for percentile 99.8 of daily mean NO_2 concentrations with the values of statistical measures are presented in Figure 11.

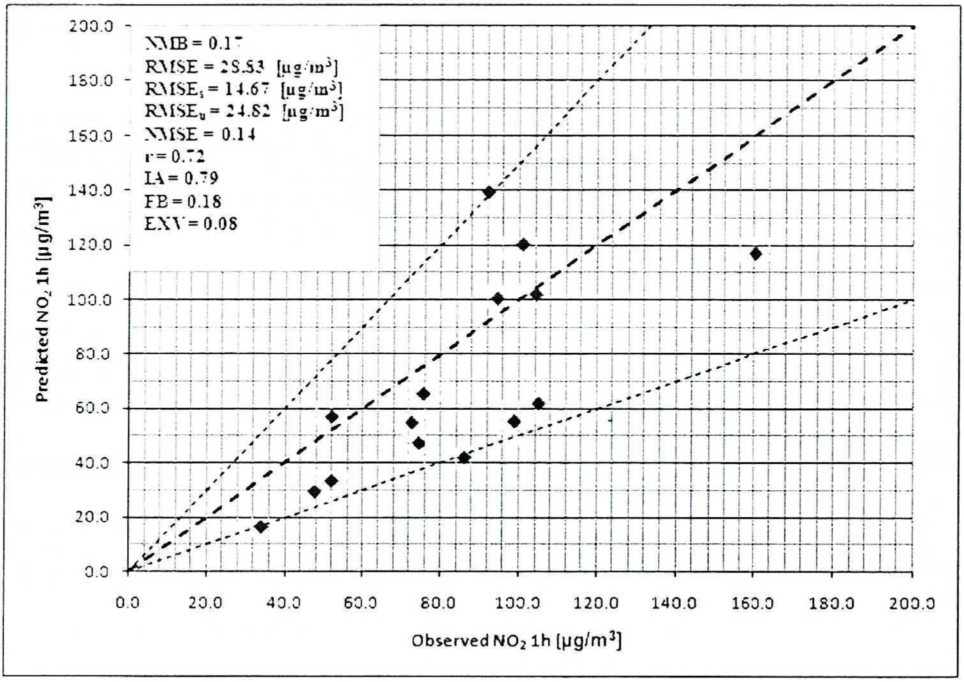

Fig. 11. Predicted and observed values for percentile 98.9 of daily mean NO_2 concentrations in [μg/m³] in Mazowieckie voivodeship; dotted lines show the variability scope imposed by the Minister's order (± 50%) and ideal conformity of the model with the measurements (the middle line)

The comparison of calculated NO_2 daily mean concentration time series with the values measured at the station in Legionowo is presented in Figure 12.

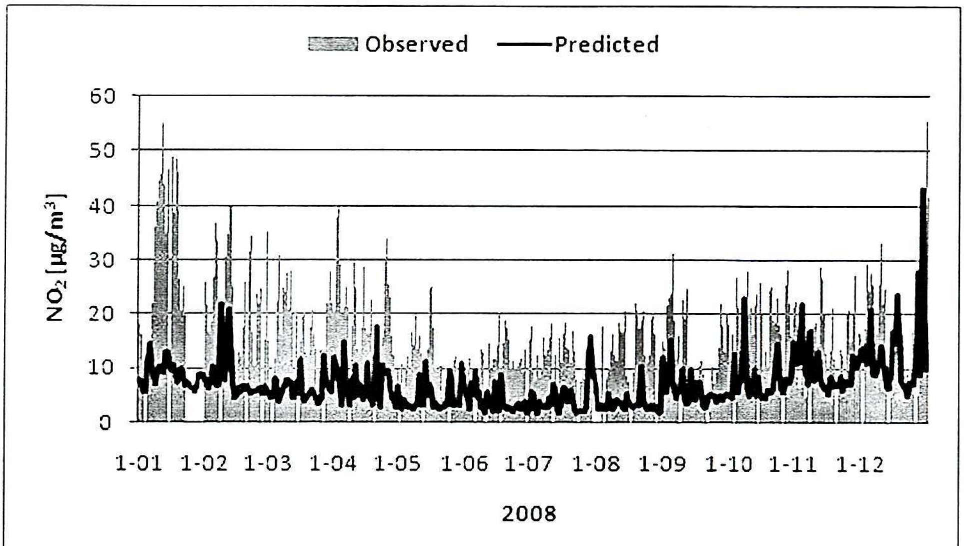

Fig. 12. Daily means of NO_2 concentrations, estimated and observed, in [μg/m³] time series in Legionowo, 2008

The obtained statistical assessment of the model operation for NO_2 (Fig. 11) indicates, that there were some problems with NO_2 modeling. Fairly high correlation and agreement indices ($r = 0.7$; $IA = 0.79$) were obtained, as well as satisfactorily low bias and error values: NMB = 0.17, FB = 0.18, NMSE = 0.14. Moreover, all the modeling results fit into the permissible variability scope imposed by the Ministry of Environment's Order for modeling results versus measurement results. However, it is clear that the dispersion of the results is definitely larger than in the case of SO_2. The value of variance explained by the model is rather unsatisfactory (EXV = 0.08). Daily concentration time series presented in Figure 12 show that the model poorly follows the observed values. As can be clearly seen, the model underestimates measuring values of NO_2, especially the high concentrations observed in January 2008. It seems that it is necessary to perform a critical review and verification of NO_2 emission inventory and indicators of NO_2 emission from individual heating and traffic that are used in Poland.

Figure 13 presents the results of PM_{10} modeling with the WRF-CALMET-CALPUFF system of models. It must be emphasized that PM_{10} is currently a priority pollutant in the EU due to the highest (apart from ozone) threat to human health. Figure 13 shows the distribution of annual mean PM_{10} concentrations whose limit value amounts to 40 μg/m³.

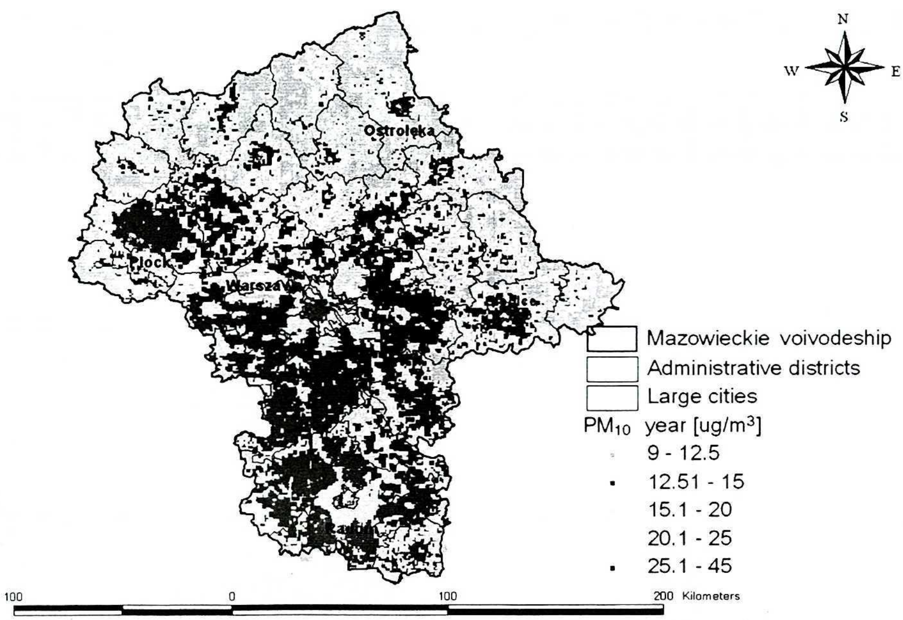

Fig. 13. PM_{10} concentrations; annual mean value expressed in [μg/m³] in Mazowieckie voivodeship, 2008

As can be seen in Figure 13, PM_{10} concentrations are the lowest (9–12.5 μg/m³) in rural areas of the northern and north-eastern part of Mazowieckie voivodeship. In the rest of the rural areas of the voivodeship the concentrations are much higher (12.5–20 μg/m³). The highest concentrations (20.1–45 μg/m³) appear in cities; e.g., in Warsaw Agglomeration a distinct area of concentrations exceeding the limit value of 40 μg/m³ can be noticed.

The scatter plot of the predicted and observed values for annual mean PM_{10} concentrations with the values of statistical measurements are presented in Figure 14.

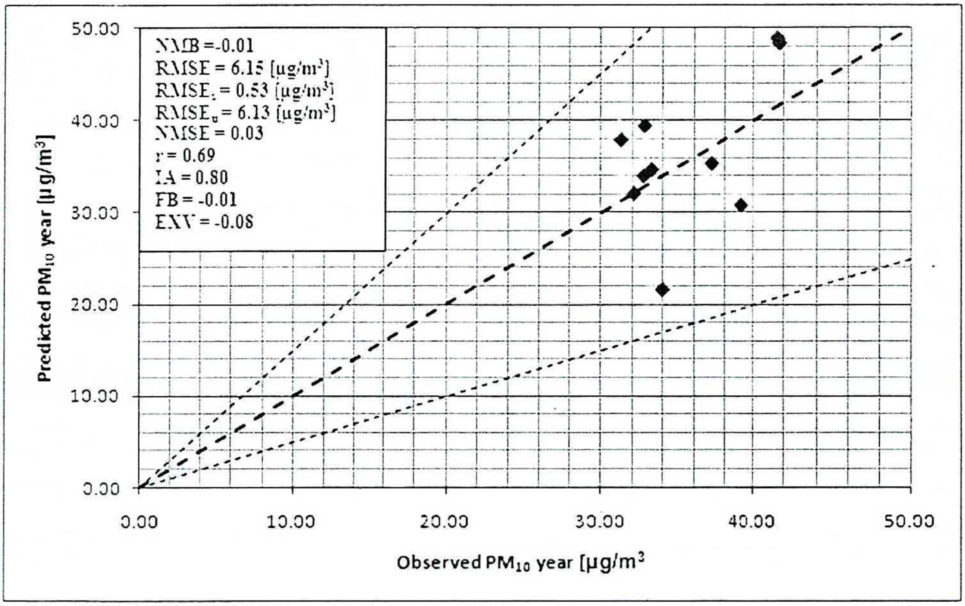

Fig. 14. Predicted and observed values for annual mean PM_{10} concentrations in [µg/m³] in Mazowieckie voivodeship; dotted lines show the variability scope imposed by the Minister's order (± 50%) and ideal conformity of the model with the measurements (the middle line)

The comparison of calculated PM_{10} daily mean concentration time series with values measured at a traffic station in Warsaw (at Niepodległości alley) is presented in Figure 15.

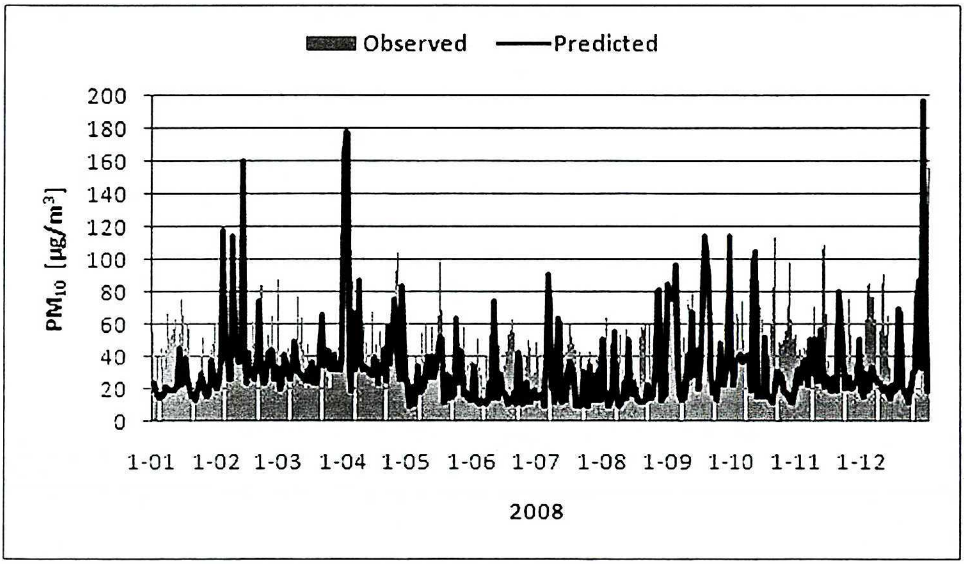

Fig. 15. PM_{10} daily mean concentrations time series, in [µg/m³], in Warsaw (Niepodległości alley), 2008

The obtained statistical measures assessing the operation of the model for PM_{10} (shown in Fig. 14) indicate that the CALPUFF model calculates PM_{10} characteristics in a very satisfactory way. Both the correlation coefficient (r = 0.69), as well as the index of agreement (IA = 0.80) prove a very strong relation between modeled and measured values. All the modeling results fit into the permissible variability scope imposed by the Ministry of Environment Order for modeling results versus measurement results and error indicators allow to state that the performance of the model is very good: NMB = 0.01, FB = 0.01, NMSE = 0.03. Only the model-explained variance (EXV = -0.08) has a low value, but it is a very "demanding" measure of model performance [13]. Time series presented in Figure 15 indicate that both in terms of values, as well as temporally, the model keeps up with observed values very well and can excellently simulate high concentration values.

The additional essential advantage of the CALPUFF model is its ability to perform separate calculations of concentrations coming from any sources and subsequently summing them up in each receptor. Consequently, it is possible to determine precisely the share of each source in the concentrations calculated in any receptor. The major share of emission types and pollution inflow in PM_{10} concentrations in Mazowieckie voivodeship in 2008 is presented in Figure 16.

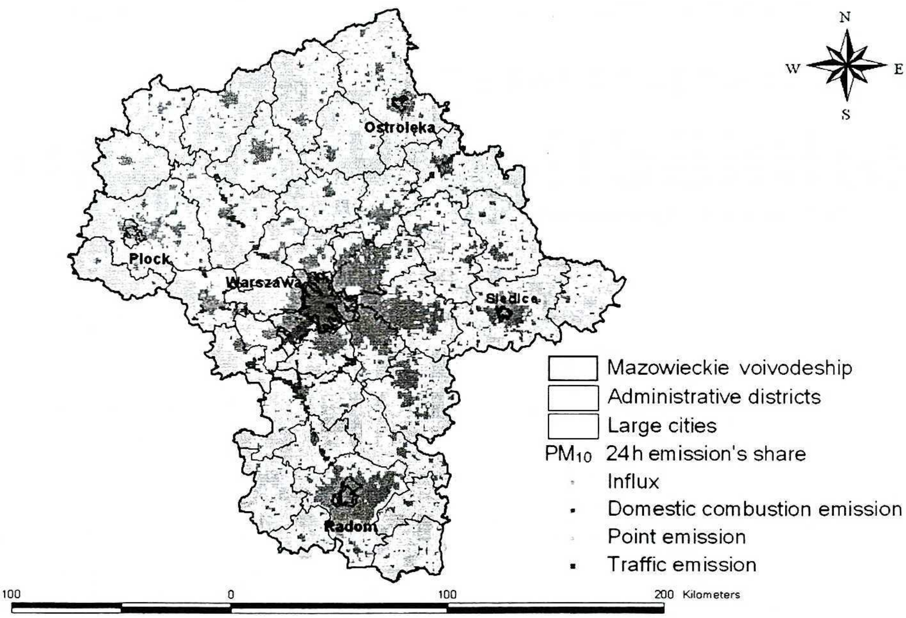

Fig. 16. Major share of emission types and pollution inflow in daily mean concentrations PM_{10}, percentile 90.4, in Mazowieckie voivodeship in 2008

As can be seen from Figure 16, in the rural area of Mazowieckie voivodeship the largest share in percentile 90.4 of daily mean PM_{10} concentrations is due to pollution inflow from the outside of the voivodeship. In Warsaw and along the largest traffic arteries traffic emission prevails, whereas in urbanized areas (zones around Warsaw and larger cities) municipal emission constitutes the major part of PM_{10} concentrations. The influence of point emission (heat and power plants, industrial sources) on the concentration of PM_{10} dust in Mazowieckie voivodeship is the least significant.

SUMMARY AND FINAL CONCLUSIONS

The study presents the calculation results of gas (SO_2 and NO_2) and dust (PM_{10}) air pollutant concentration distributions in Mazowieckie voivodeship in 2008. The calculations involved the application of WRF-CALMET-CALPUFF system of models. A very detailed emission database comprising 22 000 emitters was prepared. Pollution inflow from the outside of the area was taken into consideration as well, based on temporally and spatially variable boundary conditions calculated with the application of results of the EMEP model. Three-dimensional meteorological data were taken from the American NCEP/NCAR Reanalysis project servers.

The obtained pollution concentration distributions indicate that in Mazowieckie voivodeship atmospheric air cleanness is under the greatest threat due to dust pollution. Concentrations of the main gas pollutants that were analyzed did not exceed limit values anywhere and their concentrations did not exceed the upper threshold for estimated variables. However, PM_{10} dust concentrations did exceed annual mean and daily mean limit values, what may have caused negative health effects on the voivodeship inhabitants, especially those living in the Warsaw Agglomeration.

The assessment of the WRF-CALMET-CALPUFF system of models performance (dispersion charts of calculated versus measured values, time series comparison of calculated versus measured values, as well as the values of 9 statistical measures applied) proves the applied modeling solution to be definitely useful in application in the air quality assessment system in Poland. At the same time, the presented calculation results for NO_2 concentrations indicate the need of further work both in the area of emission (especially traffic emission) databases, as well as in the case of model implementation.

It must be especially emphasized that the CALPUFF model for dust dispersion simulation has gained a positive assessment. PM_{10} dust is, on the one hand, a priority pollutant in the EU, and on the other hand, it is a non-homogenous compound, both of primary and secondary origin, what makes it the most difficult air pollutant in terms of mathematical modeling. The results presented in this work indicate that the CALPUFF model can be successfully applied for the purposes of simulating dust dispersion in the atmosphere, and the conformity with measurement results that was obtained is better than the results of the majority of higher class models like a new generation three-dimensional Eulerian models [27].

The results presented allow us to conclude that the CALPUFF new generation Gaussian puff model, though not the easiest in operation it, can be fully used and meets all the requirements imposed by the law. It should be therefore recommended to be used in air quality assessment in Poland and in the near future it should replace the thirty-year-old Pasquill's model in Polish law.

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ZASTOSOWANIE MODELI CALMET/CALPUFF W SYSTEMIE OCENY JAKOŚCI POWIETRZA W POLSCE

Wejście Polski do Unii Europejskiej i związana z tym potrzeba prowadzenia ocen jakości powietrza atmosferycznego w strefach spowodowało radykalną zmianę wymagań wobec narzędzi stosowanych w ramach oceny. Zgodnie z polskim prawem strefą jest aglomeracja powyżej 200 000 mieszkańców oraz powiat bądź grupa powiatów, a w ocenie muszą być uwzględniane globalny i regionalny napływ zanieczyszczeń oraz wpływ lokalnych źródeł emisji oraz istotnych źródeł emisji z województwa. Tak określone wymagania narzuciły przestrzenny zasięg modelu na powyżej 250 km. Po analizie różnych pracujących na świecie systemów modeli, do wdrożenia w systemach oceny jakości powietrza w strefach w Polsce wybrano model CALPUFF z preprocesorem meteorologicznym CALMET. W pracy przedstawiono wyniki obliczeń modelowych przeprowadzonych w ramach oceny jakości powietrza w województwie mazowieckim oraz porównanie otrzymanych wyników z wynikami pomiarów na automatycznych stacjach monitoringu powietrza.