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FINE-RESOLUTION MODELING OF CONCENTRATION AND DEPOSITION OF NITROGEN AND SULPHUR COMPOUNDS FOR POLAND - APPLICATION OF THE FRAME MODEL

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Abstract: The main source of spatial information on concentration and deposition of air pollutants in Poland is the continental scale EMEP model with 50 km x 50 km grid. The coarse resolution of the EMEP model may be insufficient for regional scale studies. A new proposal is the application of the national scale atmospheric transport model FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange), originally developed for the United Kingdom. The model works with 5 km x 5 km spatial resolution and the air column is divided into 33 layers. FRAME was used here to assess the spatial patterns of yearly averaged air concentrations, and wet and dry deposition of sulphur and nitrogen compounds for the area of Poland. This study presents preliminary results of the modeling of the yearly average concentrations as well as dry and wet depositions of SO_x, NO_y and NH_x for Poland. FRAME results were compared with available measurements from the monitoring sites and national deposition budget with the EMEP and IMGW estimates. The results show close agreement with the measured concentrations expressed by determination coefficient close to 0.7 for both SO_2 and NO_x. The dry and wet deposition budgets for FRAME are also in close agreement with the EMEP and GIOŚ estimates. The FRAME model, despite its relatively simple meteorological parameterizations, is well suited to calculate the spatial pattern of annual average concentration and yearly deposition of atmospheric pollutants which was earlier presented for the UK and was shown in this paper for Poland. The model can also be used to analyze the impact of individual point sources or different emission sectors on spatial pattern of air concentration and deposition as well as testing the changes in deposition resulting from future emissions reduction scenarios.

INTRODUCTION

During the last two decades abatement strategies have been applied, in Poland and internationally, to reduce pollutants' emissions. Atmospheric long-range transport models are important instruments to estimate the fate of pollutants and to understand the effects of changes in emissions. Pollutant deposition problem is a regional issue rather than a local one with pollutants found about a thousand kilometers from the source. This implies that co-operation at an international level is necessary [3]. In Poland, main sources of air pollution are the following: energy production (contribution to the total SO_2 emission approaches 50%), industry, municipal sector and transport [10]. Over the last ten years the sulphur and nitrogen deposition have decreased in Poland as a result of decreased emissions. However, the relative contribution of ammonia deposition shows an increasing trend.

The concentrations and deposition of atmospheric pollutants are calculated across Europe with the Unified EMEP model with 50 km x 50 km grid resolution [29]. These deposition and concentration fields provide a useful guide to the magnitude of pollutant deposition and concentration, however, there is a need for individual countries to use their own national scale models to resolve atmospheric physical and chemical processes which occur at a much finer resolution than that currently available with the EMEP model. Air pollution modeling in Poland has been carried out for the last 20 years, including, among others, the urban scale SO_2 modeling [17], regional scale SO_x and NO_x modeling [1, 18], modeling for the regulatory purposes [22], the regional and long-range transport of heavy metals [4] and, recently, oxidants modeling [19].

This paper brings a general description of the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME), as well as the input data which are used for the calculation of the yearly average air concentration and deposition for the area of Poland with 5 km x 5 km grid resolution. The model was developed at the Centre for Ecology and Hydrology (CEH) Edinburgh and has been successfully used for modeling long-range transport and deposition of atmospheric pollutants in the United Kingdom. The model is also used as a tool to support government policy in assessing the effects of abatement of pollutant gas emissions and in Integrated Assessment Modeling [28] and exceedances of critical load.

This paper presents the preliminary results of the SO_x, NO_x and NH_x air concentrations, wet and dry deposition modeling for Poland. The modeling results are compared with the available measurements and with the EMEP and GIOŚ estimates of the national deposition budgets. Possible applications of the FRAME model are also discussed.

THE FRAME MODEL

A detailed description of the Fine Resolution Multi-pollutant Exchange model (FRAME) can be found in [8, 13–15, 30]. The FRAME model is a Lagrangian atmospheric transport model used to assess the annual mean air concentration and deposition of atmospheric pollutants. The model was developed from an earlier European scale model, TERN (Transport over Europe of Reduced Nitrogen [2]).

FRAME simulates an air column moving along straight-line trajectories. Trajectories are run at a 1° resolution for all grids at the edge of the model domain. The air column advection speed and frequency for a given wind direction is statistically derived from radio-sondes measurements [7]. The adoption of straight line trajectories was found to be successful in reproducing annual average measurements of gas and aerosol concentrations in air and wet deposition in the UK [8, 24, 25].

The atmosphere is divided into 33 separate layers extending from the ground to an altitude of 2500 m. Layer thicknesses vary from 1 m at the surface to 100 m at the top of the mixing layer. Vertical diffusion in the air column is calculated using K-theory eddy diffusivity and solved with the Finite Volume Method. Point source emissions are treated individually with a plume rise model. Additional information on stack height, temperature and velocity of the outflow gases are used to calculate an effective emissions height [33]. The plume reaches its maximum height when temperature is equal to the surrounding environment and its momentum is dissipated. Buoyancy forces dominate the plume rise, which is parameterized separately for stable conditions and for neutral and unstable conditions according to the Pasquill-Gifford stability classes. The depth of the boundary layer in FRAME is calculated using a mixed boundary layer model with constant potential temperature capped by an inversion layer with a discontinuity in potential temperature. Solar irradiance is calculated as a function of latitude, time of the year and time of the day. Initial gas and aerosol concentrations at the edge of the model domain are calculated with FRAME-EUROPE, a European scale model working with a 50 km x 50 km resolution.

The chemical scheme used in FRAME is similar to the one employed in the EMEP Lagrangian model [3]. The prognostic chemical variables calculated in FRAME are: NH_3, NO, NO_2, HNO_3, PAH, SO_2, H_2SO_4, as well as NH_4^+, NO_3^- and SO_4^{2-} aerosols.

Dry deposition velocities of SO_x, NO_y and NH_x are ecosystem specific and calculated individually to five different land cover categories (forest, grassland, moorland, urban and arable). For ammonia, the deposition velocity is generated from the sum of the aerodynamic resistance, the laminar boundary layer resistance and the surface resistance [30].

The model employs a constant drizzle approach using precipitation rates calculated from a climatological map of annual precipitation for Poland [20]. Wet deposition of chemical species is calculated using scavenging coefficients based on those used in the EMEP model. An enhanced washout rate is assumed over mountainous areas due to the scavenging of cloud droplets by the seeder-feeder effect to calculate local scale orographic enhancement of precipitation and concentration [6]. The washout rate for the orographic component of rainfall is assumed to be twice that calculated for the non-orographic component.

INPUT DATA

A detailed inventory of annual emissions from individual point sources (568, 742 and 43 for SO_2, NO_2 and NH_3 respectively) for the year 2004 is provided by the EPER database [11]. If available, additional information is provided to calculate the effective emission height. These include data on the stack height and diameter and temperature and exit velocity of the outflow gases.

The national totals of low level emissions of SO_2 and NO_2 from residential combustion for the area of Poland are taken from the National Inventory Report for the year 2002 [27] and disaggregated spatially with the data on fuel consumption for heating and population number provided by the National Statistical Office at the commune level [26]. The NO_2 emission from road transport is estimated using the detailed information on traffic intensity provided by the General Directorate for National Roads and Motorways. Maps of the ammonia emissions are developed using the methodology proposed by [9] and information on the animal numbers and fertilizer consumption, provided by the National Statistical Office [26]. The low-level emission of NH_3 was calculated separately for cattle, pigs, poultry and sheep [21]. This is because of the model's high vertical resolution which allows input of emissions to different heights. Aerial emissions of SO_2, NO_2 and NH_3 are calculated with 5 km x 5 km spatial resolution. The emission data for the remaining area of the FRAME model domain, covering eastern Germany, northern Czech Republic and

Slovakia and western parts of Lithuania, Belarus and Ukraine, are taken from the EMEP 50 km x 50 km expert emission inventory [32].

Wind frequency and wind speed roses employed in FRAME use 6-hourly operational radio-sonde data from the six stations located in Central Europe: Wrocław, Legionowo, Łeba (all three in Poland), Greifswald, Lindenberg (Germany), Prague, Poprad and Kiev (Czech Republic, Slovakia and Ukraine, respectively), spanning the whole 2002 year period (Fig. 1). The wind roses were developed with the methodology proposed by [7].

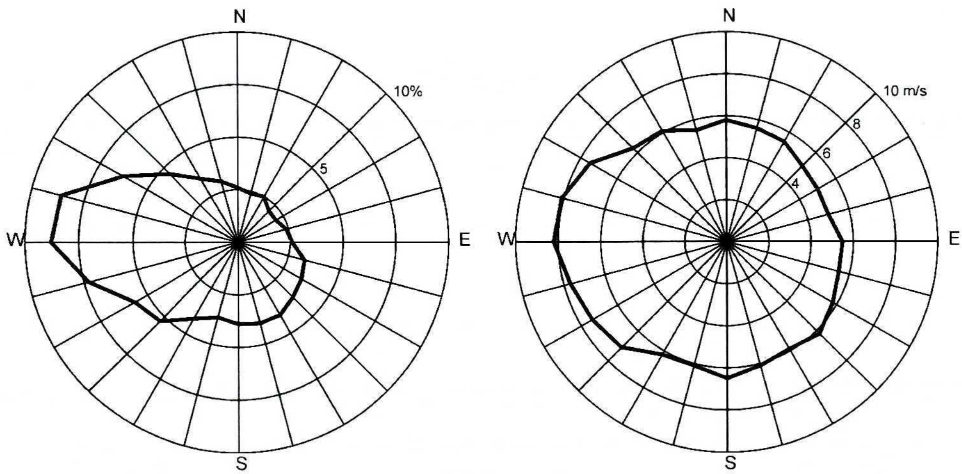

Fig. 1. Wind speed and frequency roses used in simulations (2002)

MODEL VALIDATION

The FRAME modeled yearly averaged air concentrations of SO_2 and NO_x were checked against the available measurements [16]. The data from 75 monitoring sites were available for SO_2 [31]. In the case of NO_x, the data from 27 sites were taken from the AirBase database. NH_3 concentrations were measured only on the three sites operating in the EMEP network. The NH_3 measuring sites are Jarczew (51°49'N 21°59'E), Śnieżka (50°44'N 15°44'E) and Łeba (54°45'N 17°32'E).

Based on the measured and modeled SO_2 and NO_x air concentrations, quantitative metrics are calculated to evaluate the FRAME model performance. Detailed description of the calculated statistics can be found in [12, 18, 23, 34, 35], and are summarized, after [35], in Table 1.

The predicted and measured mean concentrations are the simplest and most general measures of the model performance. MB is usually interpreted as a measure of overall under-or overestimation by the model, while MAGE and RMSE characterize the spread of the departure between the model and observations [35]. Two measures of relative difference, MNB and MNAE, are useful in comparing the performance of the model for different chemical species (SO_2 and NO_x in this case). For the FRAME model, the model-data agreement is also presented on the scatter plot, together with regression analysis and determination coefficient. The 1:1, 2:1 and 1:2 lines are shown on the scatter plots for reference.

Table 1. Quantitative metrics used in model validation (N – total number of pairs, M_i – modeled concentrations, O_i – measured concentrations)

Metrics	Mathematical expression
Measured Mean	$MM = \frac{1}{N}\sum_i O_i$
Predicted Mean	$PM = \frac{1}{N}\sum_i M_i$
Mean Bias	$MB = \frac{1}{N}\sum_i (M_i - O_i)$
Mean Absolute Gross Error	$MAGE = \frac{1}{N}\sum_i \lvert M_i - O_i \rvert$
Root Mean Squared Error	$RMSE = \sqrt{\frac{1}{N}\sum_i (M_i - O_i)^2}$
Mean Normalized Bias	$MNB = \frac{1}{N}\sum \left(\frac{M_i - O_i}{O_i} \right)$
Mean Normalized Absolute Error	$MNAE = \frac{1}{N}\sum \left(\frac{\lvert M_i - O \rvert_i}{O_i} \right)$

Despite providing the measures of the FRAME performance, the same statistics are calculated also for the EMEP model estimates. The comparison of the error statistics calculated for these two different models can answer the question if there is any gain in applying higher resolution model for regional assessment of air pollution.

FRAME calculated spatial patterns of annual average air concentration and yearly deposition (dry, wet and total) for SO_x, NO_y and NH_x are compared visually with the EMEP model results. Wet, dry and total national deposition budgets are calculated for the FRAME model and compared with the estimates presented by EMEP and GIOŚ (wet deposition only). It should be noticed that GIOŚ wet deposition budget estimates are based on 25 point measurements of ion concentrations in rainfall which are spatially interpolated to produce maps of wet deposition in Poland.

RESULTS AND DISCUSSION

The spatial patterns of yearly average ground level concentrations of SO_2, NO_x and NH_3 show general agreement with the EMEP estimates (Figs 2 to 4). The areas with the highest modeled concentrations are close to the source regions of low-level emissions for all chemical species. The main sources of low-level emissions in Poland are residential combustion, traffic and agriculture (both animal breeding and fertilizer consumption) for SO_2, NO_x and NH_3, respectively. Because of the high spatial resolution of the FRAME model, the calculated concentrations are locally significantly higher than estimated by the 50 km x 50 km EMEP model. For the EMEP model the concentrations are given for the lowest layer which is 50 m thick. For the FRAME model, air concentrations are given for the lowest layer of 1 m thickness and this can also explain the differences between the FRAME and EMEP estimates.

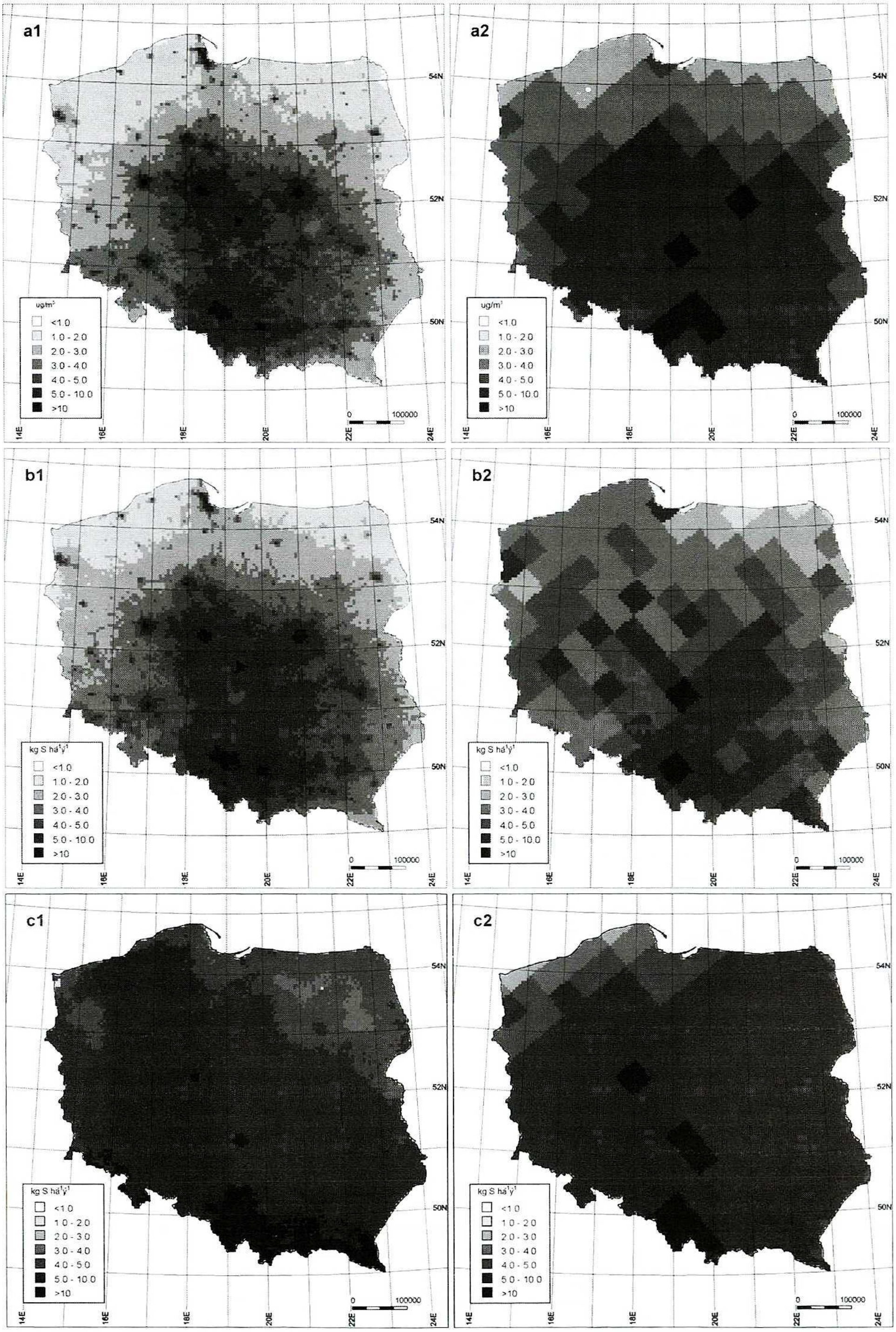

Fig. 2. FRAME (1) and EMEP (2) modeled surface concentration (a), dry (b) and wet (c) deposition of SO_x for Poland in 2002

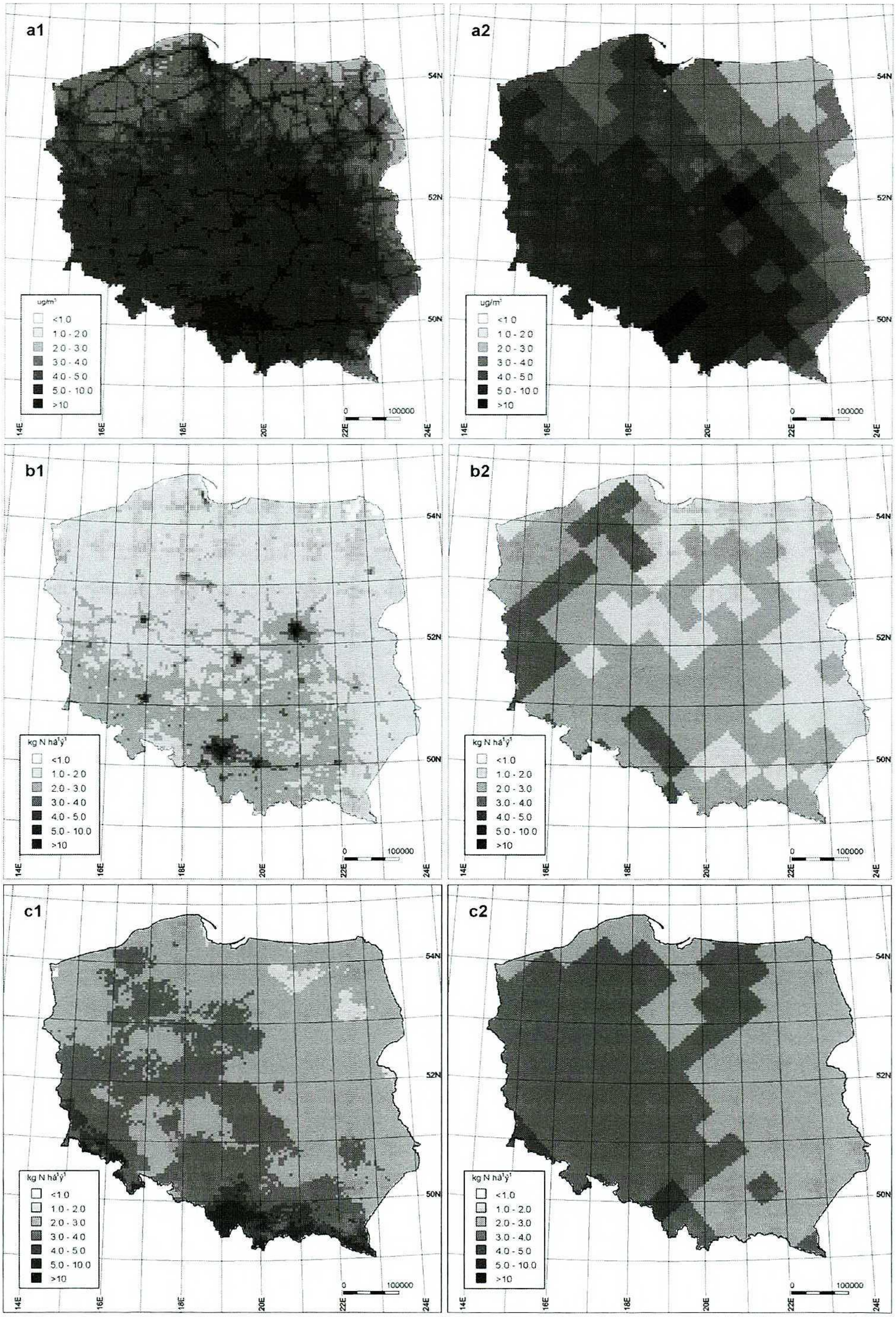

Fig. 3. FRAME (1) and EMEP (2) modeled surface concentration (a), dry (b) and wet (c) deposition of NO_y for Poland in 2002

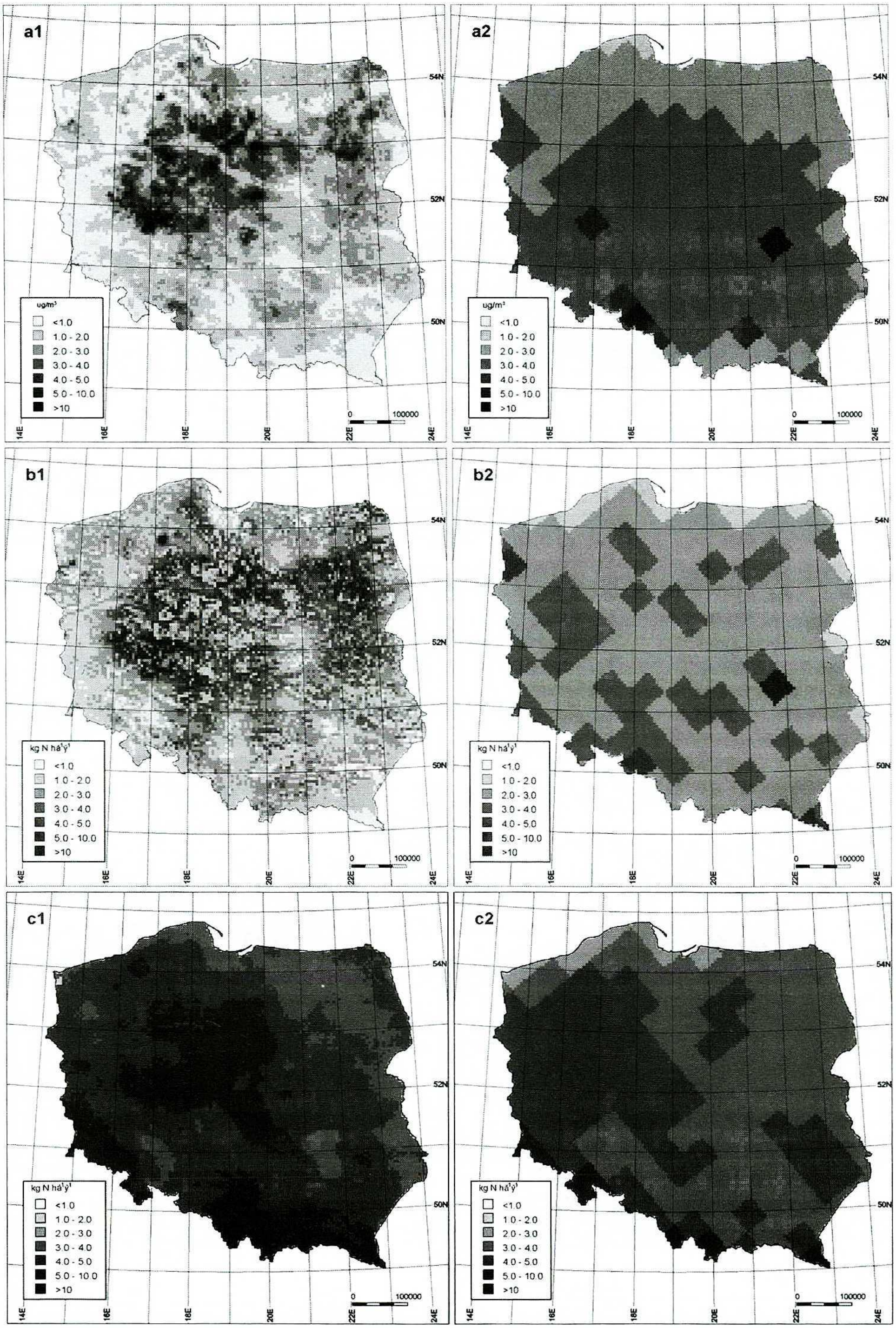

Fig. 4. FRAME (1) and EMEP (2) modeled surface concentration (a), dry (b) and wet (c) deposition of NH_x for Poland in 2002

The FRAME modeled air concentrations of SO_2 and NO_x show good agreement with the measurements collected at the GIOŚ monitoring network. The determination coefficient (R^2) for both chemical species is over 0.6 and 0.7 for SO_2 and NO_x respectively (Fig. 5). The determination coefficients are statistically significant, with p-value < 0.05. FRAME tends to strongly underestimate the concentrations on urban stations, which is clearly visible for SO_2 (triangles in Fig. 5). This is because of the large sub-grid scale variations in concentration, usually caused by local emission sources. If the measuring site is located close to the emission source, the difference between model estimates and measurements can be significant, as the model calculates average conditions in a 5 km x 5 km grid. Therefore, urban and traffic stations were excluded from the model validation subset, as being unrepresentative for the larger area.

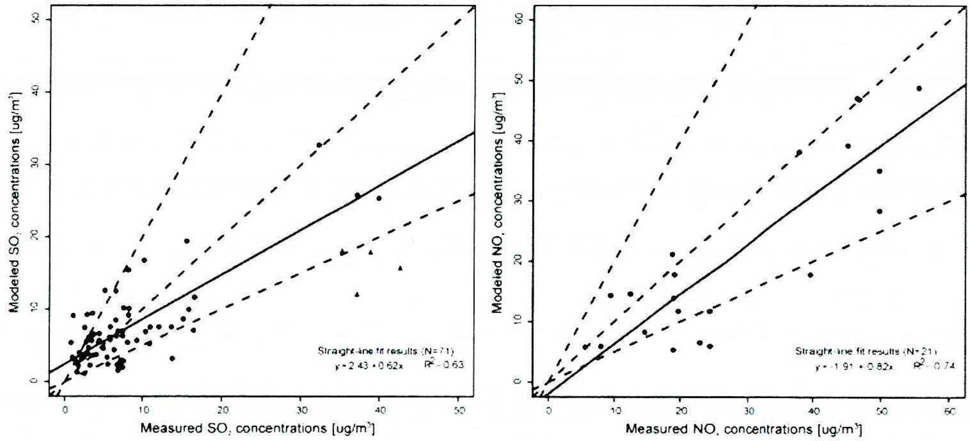

Fig. 5. SO_2 and NO_x concentrations scatter plots (triangles – urban stations, omitted in statistics calculations, solid line – best fit, dashed – reference lines 1:1, 2:1 and 1:2)

For both sulphur and nitrogen oxides, the FRAME model tends to underestimate the predicted air concentrations. This is suggested by the higher MM than PM values, as well as negative MB (Tab. 2). Despite the absolute value of MB for NO_x being 4 times larger than calculated for SO_2, relative values (MNB) suggest that the model performs better for nitrogen oxides than for SO_2. This is also supported by the MNAE. It should be however noticed, that the SO_2 statistics are based on larger number of measurements therefore such comparison is not straightforward.

Table 2. FRAME and EMEP model performance statistics

Statistics	SO_2 (N = 71)		NO_x (N = 25)	
	FRAME	EMEP	FRAME	EMEP
MM [µg/m³]	9.19		28.01	
PM [µg/m³]	7.56	5.91	21.14	7.38
MB [µg/m³]	-1.63	-3.28	-6.87	-20.63
MAGE	4.38	5.45	7.86	20.75
RMSE	6.82	9.68	10.59	24.55
MNB	0.30	0.26	-0.23	-0.65
MNAE	0.71	0.73	0.31	0.68

Almost all statistical measures used in model evaluation favor the FRAME model. The FRAME PM is closer to MM for both sulphur and nitrogen, MB is closer to zero and average errors, given by MAGE and RMSE, are smaller. These suggest that the detailed resolution of the FRAME model is very important on the air concentrations estimates, despite the simplified meteorology used for modeling. The gain is larger for NO_x than for SO_2, which may be attributed to differences in emission sources. The majority of sulphur emission comes from large point sources, while large amounts of NO_x are emitted e.g. from road transport i.e. locally and close to the ground. Therefore the EMEP model, due to its coarse spatial and vertical resolution, may not be able to describe NO_x air concentrations properly. This is of special importance in assessing the critical levels, as the underestimation of S and N concentrations will influence the final results.

NH_3 concentration is not routinely measured at the monitoring stations in Poland; therefore, it is not possible to validate the model estimates in a similar way as for SO_2 and NO_x. For the three measuring sites the results are (modeled and measured [μg NH_3]): Jarczew 2.00 and 1.38; Łeba 0.67 and 0.64; Śnieżka 0.48 i 0.24. It should be noticed that locations of the Łeba (sea shore) and Śnieżka stations (mountain peak) are not optimal for the model validation. These specific locations might explain the difference between the measured and modeled NH_3 air concentration for the Śnieżka station, where the monitoring site, 1602 m a.s.l. is located in a remote part of the grid cell.

The general patterns of FRAME modeled dry and wet depositions are similar to these calculated by the EMEP model. The largest differences are found for the dry deposition of NH_x, for which the FRAME model shows high spatial variation. The complex pattern of the FRAME modeled NH_x dry deposition is caused, at least partly, by the high horizontal and vertical resolution of the model. As in the case of modeled air concentrations, the highest dry depositions are predicted nearby the low level emission sources. Intensive agricultural production (animal breeding and fertilizer consumption) is responsible for large dry deposition of reduced nitrogen in central Poland. For NO_y and SO_x the highest dry depositions are estimated in large cities and are caused by high emission from residential combustion and traffic. Significant dry deposition of NO_y is also estimated by the FRAME model along the main roads, which is not the case of the coarser resolution EMEP model.

In the mountainous areas, FRAME shows significantly higher wet deposition of SO_x, NO_y and NH_x than the EMEP model. The FRAME estimated wet deposition patterns clearly show the role of the long-range transport of air pollutants and the main peaks are located in the Western Sudety Mts., Beskid Śląski and Żywiecki, where the anthropogenic emission is relatively small. The enhanced wet deposition over the mountainous areas is caused by high annual precipitation together with the seeder-feeder effect and was earlier reported by [6] and [5]. This seeder-feeder effect can be incorporated in the high-resolution FRAME model in contrary to the coarse resolution EMEP model. The preliminary comparisons of measured and FRAME modeled wet deposition show good agreement with the determination coefficient over 0.6 for all chemical species. There is, however, a need of further investigations and these results are not discussed here.

Dry, wet and total deposition budgets were calculated for the FRAME and EMEP models (Tab. 3). In general, all three sources are in good agreement. Dry and wet deposition estimated by the FRAME model is slightly lower than calculated by EMEP, with the exception of NH_x wet deposition. The differences in calculated budgets for the FRAME and EMEP models can be considered as small, taking into account differences in the input data, especially emission data and substantial differences in model formulations. The GIOŚ estimates show close agreement with FRAME.

Table 3. Dry, wet and total deposition of SO_x, NO_x and NH_x for Poland in 2002 estimated by FRAME, EMEP and IOŚ/IMGW [Gg of S or N]

	SO_x			NO_x			NH_x		
	dry	wet	total	dry	wet	total	dry	wet	total
FRAME	129.1	202.3	331.4	57.7	93.0	150.7	79.6	146.7	226.3
EMEP	140.4	206.7	347.0	72.2	98.2	170.4	85.9	125.1	211.1
GIOS/IMGW	–	201.9	–	–	94.4	–	–	151.3	–

SUMMARY AND CONCLUSIONS

The paper presents the Fine Resolution Atmospheric Multi-pollutant Exchange model, FRAME that can be used for simulation of the long range transport, concentration and deposition of the atmospheric pollutants in Poland. The spatial patterns of air concentrations, wet and dry depositions of sulphur and nitrogen are presented. The model was validated, based on the available air concentrations measurements and the results were compared with the EMEP model.

Despite the large differences in FRAME and EMEP model formulation; there is a general agreement in the calculated spatial patterns of air concentrations and depositions of atmospheric pollutants. High resolution of the FRAME model is, however, of great importance, for example in reduced nitrogen modeling. This is because the emissions of NH_3 may vary substantially on short distances. The same is for NO_x, which is emitted in large quantities from road transport.

Both FRAME and EMEP results were compared with the measured SO_2 and NO_x air concentrations. It was found that, in general, FRAME model performs better than EMEP, despite its simplifications in meteorology. The gain is especially large for NO_x.

FRAME model tends to underestimate the sulphur and nitrogen oxides air concentrations. This issue should be further investigated. The improved emission inventory and better parameterization of the model may further improve the results.

In the mountainous areas, FRAME shows significantly higher wet deposition of sulphur and nitrogen compounds than the EMEP model. The main reason for this is orographic enhancement (seeder-feeder effect) of ion concentrations in precipitation implemented in the high resolution FRAME model, and certain difficulties related with meteorological modeling of orographic precipitation at a 50 km x 50 km resolution.

The spatial patterns of yearly averaged concentrations and deposition, both dry and wet, were presented here and checked against available measurements and estimates of other models. The close agreement is encouraging, considering the fact that the presented results are still preliminary. The improved emission inventory and better parameterization of the model may further benefit the results. Further studies should be also focused on validation of the wet deposition estimated by the FRAME model, as there are measurements that could be used for this purpose.

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MODELOWANIE KONCENTRACJI I DEPOZYCJI ZANIECZYSZCZEŃ ATMOSFERYCZNYCH W WYSOKIEJ ROZDZIELCZOŚCI PRZESTRZENNEJ – ZASTOSOWANIE MODELU FRAME

W opracowaniach dotyczących koncentracji i depozycji zanieczyszczeń atmosferycznych w Polsce, podstawowym źródłem informacji przestrzennej jest model EMEP. Jest on cennym narzędziem pozwalającym na ilościowe i jakościowe zobrazowanie przestrzennych zmian koncentracji i depozycji zanieczyszczeń oraz na szacowanie roli transportu dalekiego zasięgu w skali kontynentalnej. Jego najistotniejszą wadą jest mała rozdzielczość przestrzenna (50 km x 50 km), która ogranicza możliwości uwzględnienia procesów atmosferycznych zachodzących w skali regionalnej (np. powiązanych z rzeźbą terenu). Jednym z kilku stosowanych w Europie regionalnych modeli o wyższej rozdzielczości jest brytyjski model FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange). Wszystkie uwzględnione w nim procesy atmosferyczne i chemiczne analizowane są w kolumnie powietrza o podstawie 5 km x 5 km, podzielonej w pionie na 33 warstwy. Uzyskane za pomocą polskiej wersji modelu FRAME rozkłady przestrzenne koncentracji oraz depozycji zanieczyszczeń dla Polski dla 2002 r. charakteryzują się dobrą zgodnością z danymi pomiarowymi. W przypadku koncentracji współczynnik determinacji jest na poziomie 0,7 dla SO_2 oraz NO_2. Roczny bilans suchej oraz mokrej depozycji, wyliczony w oparciu o model FRAME, jest bliski szacunkom modelu EMEP oraz GIOŚ. Pomimo dość prostej parametryzacji danych meteorologicznych model FRAME z dobrym przybliżeniem oszacował średnią roczną koncentrację oraz roczną depozycję zanieczyszczeń. Wcześniej podobne wyniki otrzymano także dla Wielkiej Brytanii. FRAME może być więc traktowany jako użyteczne narzędzie pozwalające na przestrzenną charakterystykę średniej rocznej koncentracji i rocznej depozycji zanieczyszczeń atmosferycznych w stosunkowo wysokiej rozdzielczości przestrzennej. Model pozwala także analizować zakres oddziaływania pojedynczych źródeł emisji, czy też wpływ na środowisko poszczególnych sektorów emisji (np. osobno emisji niskiej bądź wysokiej).