Pure Appl. Geophys.

DOI 10.1007/s00024-016-1267-2

Pure and Applied Geophysics

Aerosol-Radiation Feedback and PM_{10} Air Concentrations Over Poland

Małgorzata Werner,[1,2] Maciej Kryza,[1] Carsten Ambelas Skjøth,[2] Kinga Wałaszek,[1] Anthony J. Dore,[3] Hanna Ojrzyńska,[1] and Jan Kaplon[4]

Abstract—We have implemented the WRF-Chem model version 3.5 over Poland to quantify the direct and indirect feedback effects of aerosols on simulated meteorology and aerosol concentrations. Observations were compared with results from three simulations at high spatial resolutions of 5 × 5 km: (1) BASE—without any aerosol feedback effects; (2) DIR—with direct aerosol-radiative effects (3) INDIR—with direct and indirect aerosol-radiative effects. We study the overall effect during January 2011 as well as selected episodes of the highest differences in PM_{10} concentrations between the three simulations. For the DIR simulation, the decrease in monthly mean incoming solar radiation (SWDOWN) appears for the entire study area. It changes geographically, from about −8.0 to −2.0 W m^{-2}, respectively for the southern and northern parts of the country. The highest changes do not correspond to the highest PM_{10} concentration. Due to the solar radiation changes, the surface mean monthly temperature (T2) decreases for 96 % of the area of Poland, but not more than 1.0 °C. Monthly mean PBLH changes by more than ±5 m for 53 % of the domain. Locally the differences in PBLH between the DIR and BASE are higher than ± 20 m. Due to the direct effect, for 84 % of the domain, the mean monthly PM_{10} concentrations increase by up to 1.9 μg m^{-3}. For the INDIR simulation the spatial distribution of changes in incoming solar radiation as well as air temperature is similar to the DIR simulation. The decrease of SWDOWN is noticed for the entire domain and for 23 % of the domain is higher than −5.0 W m^{-2}. The absolute differences of PBLH are slightly higher for INDIR than DIR but similarly distributed spatially. For daily episodes, the differences between the simulations are higher, both for meteorology and PM_{10} concentrations, and the pattern of changes is usually more complex. The results indicate the potential importance of the aerosol feedback effects on modelled meteorology and PM_{10} concentrations.

Key words: WRF-Chem, feedback, aerosol, PM10, Poland.

1. Introduction

Aerosol particles have an important role in the climate system acting on the global radiation budget in two ways—directly by scattering and absorbing the incoming radiation or indirectly by altering the cloud properties (e.g. Charlson *et al.* 1992; Andreae *et al.* 2005; Rosenfeld *et al.* 2008). Furthermore, a climate-biosphere feedback mechanism on diffuse radiation can alter net ecosystem exchange significantly (Mercado *et al.* 2009) and it has been suggested that this process will also be very important on the emission of particle precursors such as isoprene from nature (Wilton *et al.* 2011). Additionally, studies of human health indicated that there are significant correlations between particulate matter levels and increased respiratory and cardiovascular diseases, and mortality (Pope *et al.* 2002; Perez *et al.* 2008). Many chemical transport models (CTMs) have been developed to better understand the physical and chemical processes of gas-phase species and particulate matter. The models generally underestimate $PM_{2.5}$ and PM_{10} mass concentrations by 4.0–14.0 μg m^{-3} (10–50 %) and 6.5–18.0 μg m^{-3} (20–50 %), respectively (Tuccella *et al.* 2012). It is, therefore, important to explore the processes that relate to PM concentrations to explain this underestimation.

Most CTMs are implemented as offline models, where the meteorological input data are provided by

Electronic supplementary material The online version of this article (doi:10.1007/s00024-016-1267-2) contains supplementary material, which is available to authorized users.

[1] Department of Climatology and Atmosphere Protection, University of Wrocław, ul. Kosiby 8, 51-521 Wrocław, Poland. E-mail: malgorzata.werner@uwr.edu.pl; maciej.kryza@uwr.edu.pl; kinga.walaszek@uwr.edu.pl; hanna.ojrzynska@uwr.edu.pl

[2] National Pollen and Aerobiology Research Unit, Institute of Science and the Environment, University of Worcester, Henwick Grove, Worcester WR2 6AJ, UK. E-mail: c.skjoth@worc.ac.uk

[3] Centre for Ecology and Hydrology, Bush Estate, Penicuik, Midlothian EH26 OQB, UK. E-mail: todo@ceh.ac.uk

[4] Department of Satellite Geodesy, Wroclaw University of Environmental and Life Sciences, ul. Grunwaldzka 53, 50-357 Wrocław, Poland. E-mail: jan.kaplon@igig.up.wroc.pl

Published online: 16 March 2016

an independent model. In that solution it is impossible to simulate the complexity of the aerosol-cloud-radiation feedback process. Additionally, the decoupling between the meteorological and chemical model leads to a loss of information because of the physical and chemical processes occurring on a time scale smaller than the output time step of the meteorological data (Zhang 2008). It is well accepted that weather has a profound impact on air quality as well as that atmospheric composition can influence both weather and climate. Coupling of atmospheric dynamics, pollutant transport, chemical reactions and atmospheric composition will remain one of the most challenging tasks over the next decades as they are strongly integrated processes (Jacobson 2002; Zhang 2008; Baklanov *et al.* 2014).

Accurately simulating these feedbacks requires the use of online-coupled meteorology-chemistry models, e.g. GATOR-MMTD (Jacobson *et al.* 1996), WRF-Chem (Grell *et al.* 2005), GEM-AQ (Kaminski *et al.* 2007), GEM-MACH (Moran *et al.* 2010), among which the weather research and forecasting with chemistry (WRF-Chem) model represents a state-of-the-science online model. Zhang (2008) applied WRF-Chem over eastern Texas and showed that the presence of aerosols leads to a decrease in surface temperature by up to 0.18 °C. By coupling a cloud microphysics module with WRF, Lynn *et al.* (2007) illustrated the suppression of precipitation by continental aerosol in the Sierra Nevada Mountains. Zhang *et al.* (2010) applied WRF-Chem over North America at a 36 km × 36 km resolution to examine the influence of direct and indirect feedback effects on meteorology and photolysis rate. Despite the relatively coarse resolution, the results of Zhang *et al.* (2010) indicated the potential importance of the aerosol feedbacks on a regional scale, even at a time scale of a month. Similar studies on feedback effects have been undertaken for Europe by Forkel *et al.* (2012), where the WRF-Chem model was implemented at a resolution of 22.5 × 22.5 km. This work has shown that over the European continent, many of the spatial changes in meteorological parameters and pollutants due to aerosol effects are not only a general feature but also a result of the prevailing meteorological situation. It was suggested that a more pronounced feedback mechanism from aerosols can be expected with increased horizontal resolution (e.g. 5 vs. 50 km) or by focusing on episodes compared to long term means.

In this study we explored the direct and indirect feedback effects of aerosols on both meteorology and PM concentrations. We focused on the difference between specific episodes and long-term means and

Table 1

Model components and configuration

Category	D01	D02	D03
Simulation period	01–30 of January 2011		
Domains	Europe	Central Europe	Poland
Horizontal resolutions	45 km	15 km	5 km
Vertical resolution	35 layers		
Shortwave and longwave radiation	RRTMG		
Land-surface model	Noah LSM		
Boundary layer scheme	YSU		
Cumulus parameterization	Grell and Denvenyi (2002)	Grell and Denvenyi (2002)	Explicitly resolved
Microphysics	Lin *et al.* (1983)		
Prognostic cloud droplet number	Prognostic equation used only for the INDIR simulation		
Analysis nudging (FDDA)	Yes	Yes	No
Gas-phase mechanism	RADM2		
Aerosol model	MADE/SORGAM		
Photolysis scheme	Fast-J		
Wet deposition	Simplified parameterisation for wet scavenging		
Sea salt parameterisation	Yes (MADE/SORGAM sea salt emission)		

Please refer to the WRF and the WRF-Chem user's guides for a complete description of the options

Table 2

Mean spatial error statistics (58 stations, January 2011) for meteorological surface variables (T2, RH2, W10) for the BASE, DIR and INDIR simulations

	T2			RH2			PSFC			W10		
	BASE	DIR	INDIR	BASE	DIR	INDIR	BASE	DIR	INDIR	BASE	DIR	INDIR
MB	−2.081	−2.224	−2.226	3.858	4.013	4.032	1.539	1.555	1.564	0.593	0.587	0.589
MGE	2.540	2.637	2.636	6.925	6.937	6.918	4.067	4.070	4.074	1.504	1.503	1.507
NMB	−16.356	−18.095	−18.186	0.044	0.046	0.046	0.002	0.002	0.002	0.264	0.262	0.262
NMGE	17.576	18.879	18.953	0.078	0.078	0.078	0.004	0.004	0.004	0.485	0.485	0.486
RMSE	3.168	3.275	3.275	9.671	9.694	9.678	4.151	4.153	4.158	1.896	1.896	1.898
IOA	0.853	0.848	0.848	0.612	0.609	0.610	0.907	0.907	0.907	0.787	0.787	0.787

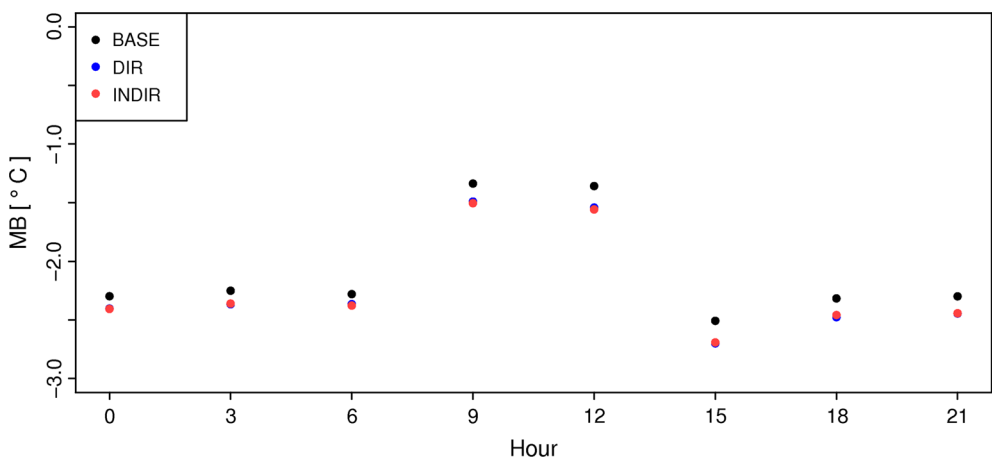


Figure 1
Diurnal cycle in mean bias for T2 for Poland for January 2012

Table 3

Error statistics for SWDOWN for Warszawa, Sopot and Strzyżów

	Warszawa			Sopot			Strzyżów		INDIR
	BASE	DIR	INDIR	BASE	DIR	INDIR	BASE	DIR	
MB	67.470	58.178	57.522	31.435	27.587	27.379	55.318	42.491	43.842
MGE	73.233	64.354	64.408	44.987	41.191	42.099	71.316	61.163	61.003
NMB	1.703	1.468	1.452	0.495	0.434	0.431	0.652	0.500	0.516
NMGE	1.848	1.624	1.626	0.708	0.648	0.663	0.840	0.720	0.719
RMSE	108.739	95.871	96.049	62.713	57.626	59.870	101.156	85.810	85.662
IOA	0.510	0.551	0.548	0.854	0.870	0.860	0.780	0.821	0.824

implemented for this purpose the WRF-Chem model version 3.5 at high spatial resolution (5 km × 5 km) over Poland. With this we studied an extended period that contains several episodes of high measured PM10 concentrations: January 2011. The aim of this study was twofold: first we wanted to compare the model results of meteorological variables and PM_{10} concentrations with available measurements. Second,

we quantified the feedback effects on modelled aerosol concentrations and meteorological parameters.

2. Methodology

2.1. WRF-Chem Setup

The Weather Research and Forecasting (WRF) model is a mesoscale non-hydrostatic meteorological model that includes a large number of options. These options include parameterisations of the Planetary Boundary Layer, the land surface description, cloud microphysics, radiation and convection processes. WRF-Chem is a version of WRF coupled online with a chemistry model where meteorological and chemical components of the model are predicted simultaneously. A complete description of the model is given by Grell *et al.* (2005) and Fast *et al.* (2006). The main options for physical and chemical schemes used here are listed in Table 1. These include the Noah Land Surface Model (Chen and Dudhia 2001), YSU boundary layer physics (Hong *et al.* 2006), RRTMG long- and short-wave radiation scheme (Iacono *et al.* 2008), Grell 3D parameterisation with radiative feedback and shallow convection (Grell 2002), the Lin microphysics scheme (Lin *et al.* 1983). The convection was explicitly resolved for the innermost domain (d03), which is of the main focus of this paper, and no analysis nudging (FDDA) was included for this domain. For the BASE and DIR simulation we used the Lin *et al.* (1983) scheme with the prognostic cloud droplet number turned off. In the INDIR simulation the cloud droplet number of grid scale clouds was calculated by a prognostic equation.

The gas phase chemistry model used in this study was the regional acid deposition model, version 2 (RADM2, Stockwell *et al.* 1990). The aerosol module included the Modal Aerosol Dynamics Model for Europe (MADE, Ackermann *et al.* 1998) for the inorganic fraction and the Secondary Organic Aerosol Model (SORGAM, Schell *et al.* 2001) for the carbonaceous secondary fraction.

The model was run for January 2011 with three one-way nested domains. This study focuses on domain 3, which covers Poland at 5 km × 5 km spatial resolution and hourly temporal resolution. The simulations were driven by the NCEP final analysis, available every 6 h, with 1° × 1° spatial resolution and TNO MACC II emissions, with 1/8° × 1/16° spatial resolution (Kuenen *et al.* 2014). Temporal variations in emissions are restricted to emissions from nature, while the TNO MACC II emissions are assumed constant during the entire simulation. The chemical boundary conditions of trace gases consist of idealised, northern hemispheric, mid-latitude, clean environmental profiles based upon the results from the NOAA Aeronomy Lab Regional Oxidant Model (Liu *et al.* 1996). The simulation uses a spin up, with the model simulation started on 30 December 2010. To study the influence of the feedback effects, we run three simulations: (1) BASE—baseline simulation, without any aerosol feedback effects; (2) DIR—direct aerosol-radiative effects only (also includes semi-direct effects); (3) INDIR—direct aerosol-radiative effects and indirect effects (also includes semi-direct and second indirect effects).

2.2. Evaluation of the WRF-Chem Model Results

The WRF modelled air temperature at 2 m (T2), relative humidity at 2 m (RH2), surface pressure (PSFC) and wind speed at 10 m (W10) were compared with 3-hourly measurements from 58 sites provided by the Institute of Meteorology and Water Management in Poland. The model evaluation was done for all three simulations—BASE, DIR and INDIR. The following statistics were calculated for all available stations as mean values for January 2011: mean bias (MB), mean gross error (MGE), normalised mean bias (NMB), normalised mean gross error, root mean square error (RMSE) and index of agreement (IOA).

Modelled downward short wave flux at ground surface (SWDOWN) was compared with measurements from three stations under the Poland-AOD network (Warszawa, Sopot, Strzyżów) and provided by the Institute of Geophysics, University of Warsaw. For Warszawa and Strzyżów the data were available for the entire analysed period and for Sopot for the first 18 days of January. The time series of modelled and observed values were provided for all stations and individual statistics for daylight hours for each station were calculated. Additionally we used all

stations from the World Radiation Data Centre available for Poland for January 2011. This included three stations (Kołobrzeg, Belsk and Zakopane) available at daily temporal resolution. For these stations we plotted time series with measured and aggregated to daily modelled values SWDOWN.

Measured hourly PM_{10} concentrations were provided by the Chief Inspectorate of Environmental Protection in Poland. 55 stations with data availability above 75 % were used to validate the modelled results. First, the average statistics, for the entire domain (FAC2, MB, RMSE, IOA) for all three simulations were calculated and presented in a table and a mean scatter plot was plotted. Then, for three sites located in the large Polish cities (Warszawa, Poznań, Łódź, marked in Fig. 8), time series of modelled and observed data were plotted and individual scatter plots presented. To check the importance of station location on model performance we used additional information on station types and plotted three Taylor diagrams (for the INDIR simulation):

1. according to type of station (background, industrial and traffic)
2. according to station type of area (rural, suburban and rural)
3. using only background stations and plotted according to station area type.

Finally the spatial distribution of MB between the INDIR simulation and observations is presented.

3. Results

3.1. Meteorology

The summary of domain-wide error statistics for all sites and the entire period of January 2011 is presented in Table 2. For all the simulations (BASE, DIR, INDIR) the lowest NMB and NMGE is for PSFC and the highest for T2. A high Index of Agreement occurs for all meteorological parameters, with only RH2 below 0.70. Inclusion of feedback effects slightly changes the statistics for T2, RH2, PSFC and W10. The highest decrease in model performance between BASE and INDIR simulation is for the air temperature. The mean bias of T2 was plotted at 3-hourly temporal resolution (Fig. 1). MB changes during the day but for all the simulations the lowest bias (below 1.5 °C) is at 9 and 12 am, whereas for the rest hours is above 2.0 °C. These diurnal changes are consistent with results reported by Kryza *et al.* (2015) for a long-term WRF simulation for Poland for years 1981–2010.

There is a reasonably good Index of Agreement (above 0.75) between modelled and observed solar radiation (SWDOWN) for Sopot and Strzyżów but observed values are overestimated by the model (Table 3; Fig. 2). The best performance has been obtained for the station located at the sea coast (Sopot) and the worse for Warszawa. Inclusion of the direct feedback improves all error statistics for the three sites; however, inclusion of indirect feedback increases MGE and RMSE for two of them, if compared to DIR. Time series plotted for daily values available from the World Radiation Data Centre for Belsk, Kołobrzeg and Zakopane (Fig. 1 in supplementary materials) present a similar trend as for hourly measurements from POLAND-AOD. The measurements are overestimated by the model, with the best agreement between model and observations for the sea cost station (Kołobrzeg) and worst for Zakopane (at the base of the mountains).

Temporal changes in SWDOWN, T2, PBLH and differences for the BASE, DIR and INDIR simulations are described for two locations—Łódź and Łeba (Fig. 3, figures for WSPD and RH2 are available in supplementary materials, Fig. 2). The first station is in the area with the highest positive differences of PM_{10} concentrations between INDIR and BASE, and the second is located in the area of the negative differences. Generally, the peak values of SWDOWN are highest for BASE and appear at the same time for all simulations. An exception is, e.g., the 12th and 17th January in Łódź, where the highest solar radiation is for INDIR and DIR, respectively. The highest differences in T2 appear for the same episodes as for SWDOWN. For selected periods the temperature in Łódź, for the INDIR simulation is up to 2.5 °C lower than that for BASE. High variability between DIR and INDIR simulations appears on 02nd–04th of January at Łódź station and 10–14th, 10–24th of January at Łeba. For several episodes

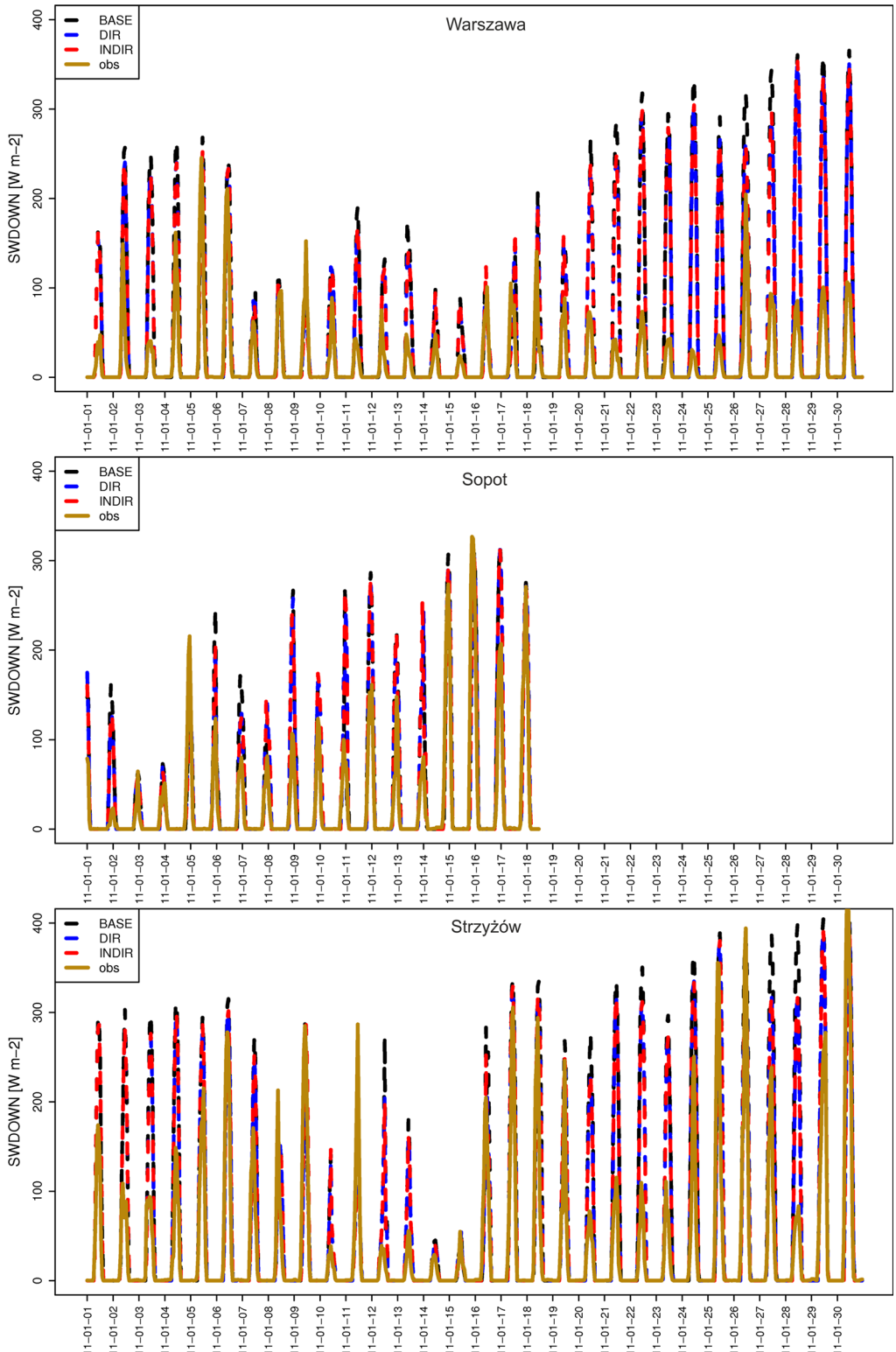


Figure 2
Modelled vs. observed hourly variation of SWDOWN

PBLH differs between DIR and INDIR simulation by more than 200 m. For Łódź, the lowest differences between the simulations are for the 4–9th and 11–15th of January, which also coincide with small differences in the air temperature. This is also a period with relatively lower values of solar radiation at the surface in comparison to the last 10 days of the month.

Mean monthly solar radiation for the BASE simulation for January 2011 in Poland domain was 53.3 W m^{-2}. Inclusion of direct effects in the model decreases the monthly mean solar radiation by 3.5 W m^{-2}. It changes geographically from about −8.0 to −2.0 W m^{-2}, respectively, for the southern and northern parts of the country. The lowest decrease is for the Baltic Sea. Solar radiation decrease is observed for the entire domain and it is more than −5.0 W m^{-2} for about 15 % of the area (Table 4). The highest changes do not correspond to the highest surface PM_{10} concentrations (Figs. 4, 9). Daily mean solar radiation changes from about −20.0 up to 10.0 W m^{-2}, both for 19th and 29th of January (Fig. 5). For the 19th the increase in SWDOWN was noticed for about 24 % of the domain. Inclusion of both direct and indirect feedback causes a decrease in monthly mean solar radiation equal to 3.8 W m^{-2}. The spatial distribution of changes are similar to changes for DIR, both for monthly and daily values (Figs. 4, 5). The decrease is apparent for the entire domain and for 23 % of the domain it is higher than 5.0 W m^{-2}. Due to the solar radiation changes, the surface mean monthly temperature (T2) decreases

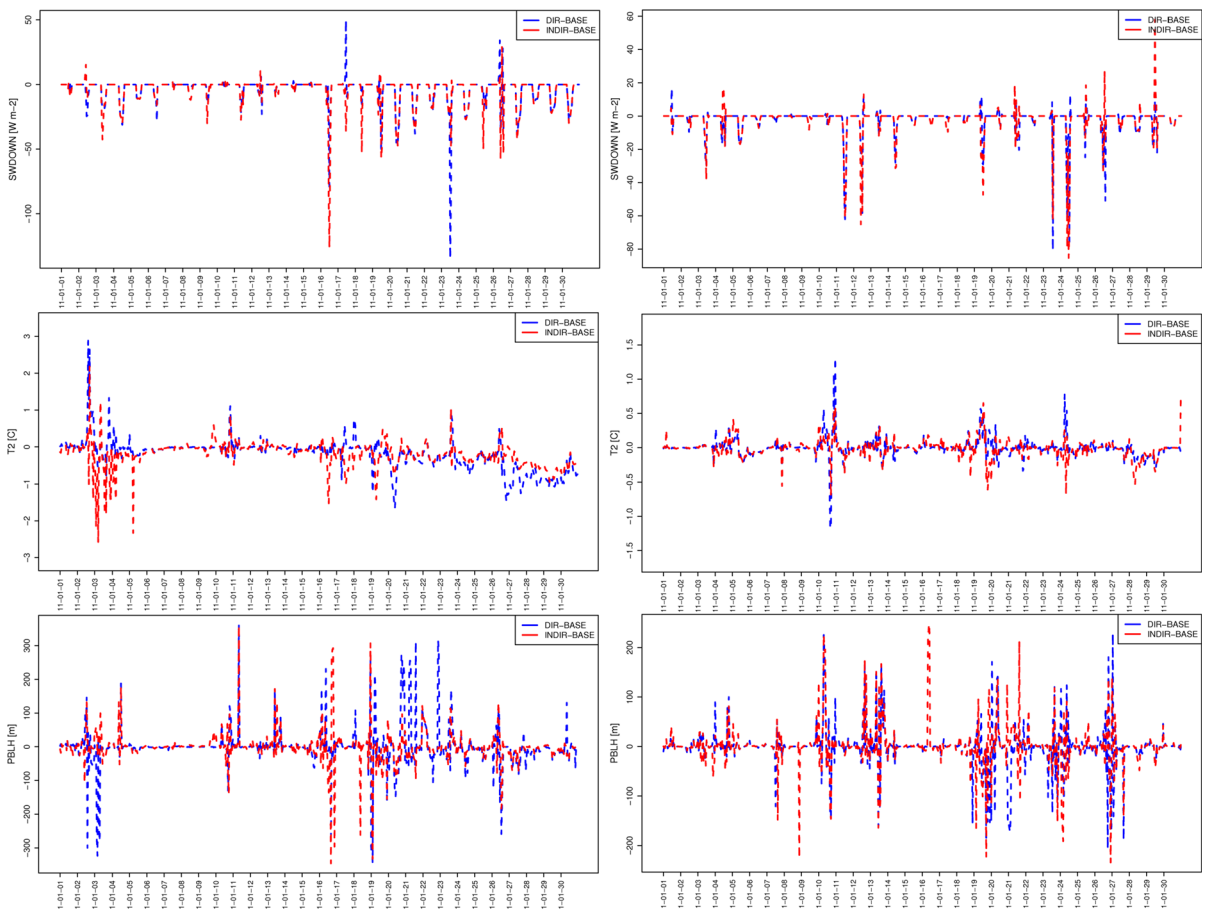


Figure 3
Hourly time series of differences in SWDOWN, T2 and PBLH between the DIR and BASE and INDIR and BASE simulations for two selected locations (*left*: Łódź, *right*: Łeba)

over 96 % of the domain, but the decrease is less than 1.0 °C (Table 4). Locally, in the north part of the country an increase was observed, of up to 0.5 °C. Daily variability is higher than monthly and for the 19th January varies between −1.5 and 2.0 °C, but for the majority of the area (about 75 %) it is negative. There are no significant differences between the results for the INDIR and DIR simulations (Table 4).

Mean PBLH for the BASE simulation in January 2011 was 243.3 m and varied from 62 to 502 m. The highest was over sea and the lowest in the south-eastern Poland. PBLH changes by more than ±5 m for 42 % of the domain when the direct feedback effect was included. The highest differences, both in the case of the DIR and INDIR simulations, were modelled for northern and southern Poland. For the northern part of the domain, DIR gives higher PBL than the BASE simulation up to 14 m, whereas for the southern part PBLH is lower for DIR by up to 23 m. For the episodes of the 19th and 29th January the differences exceed 20 m, respectively, for about 33 and 10 % of the domain; however, for some regions the difference is above 50 m and locally even exceeds 100 m.

Figure 4
Monthly mean spatial differences in SWDOWN, T2 and PBLH between DIR and BASE (*left column*) and INDIR and BASE (*right column*)

3.2. PM_{10} Concentrations

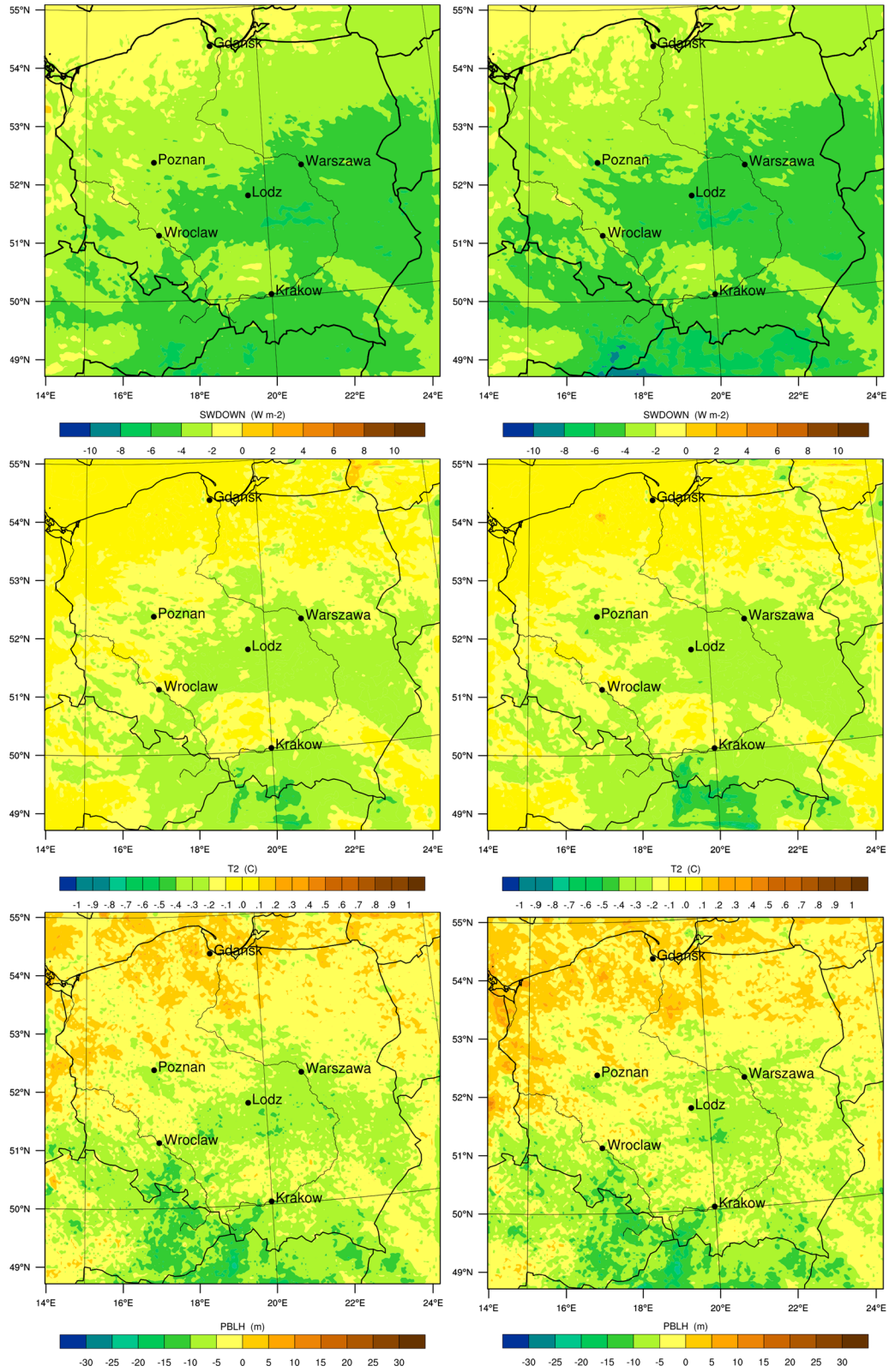
The model results for all three simulations have been compared with the surface PM_{10} measurements. There are no large differences in mean model performance among the simulations (Table 5; Fig. 6). For all of the three runs the FAC2 statistic is 0.89, MB equal from −9.54 to −9.70 μg m^{-3} and

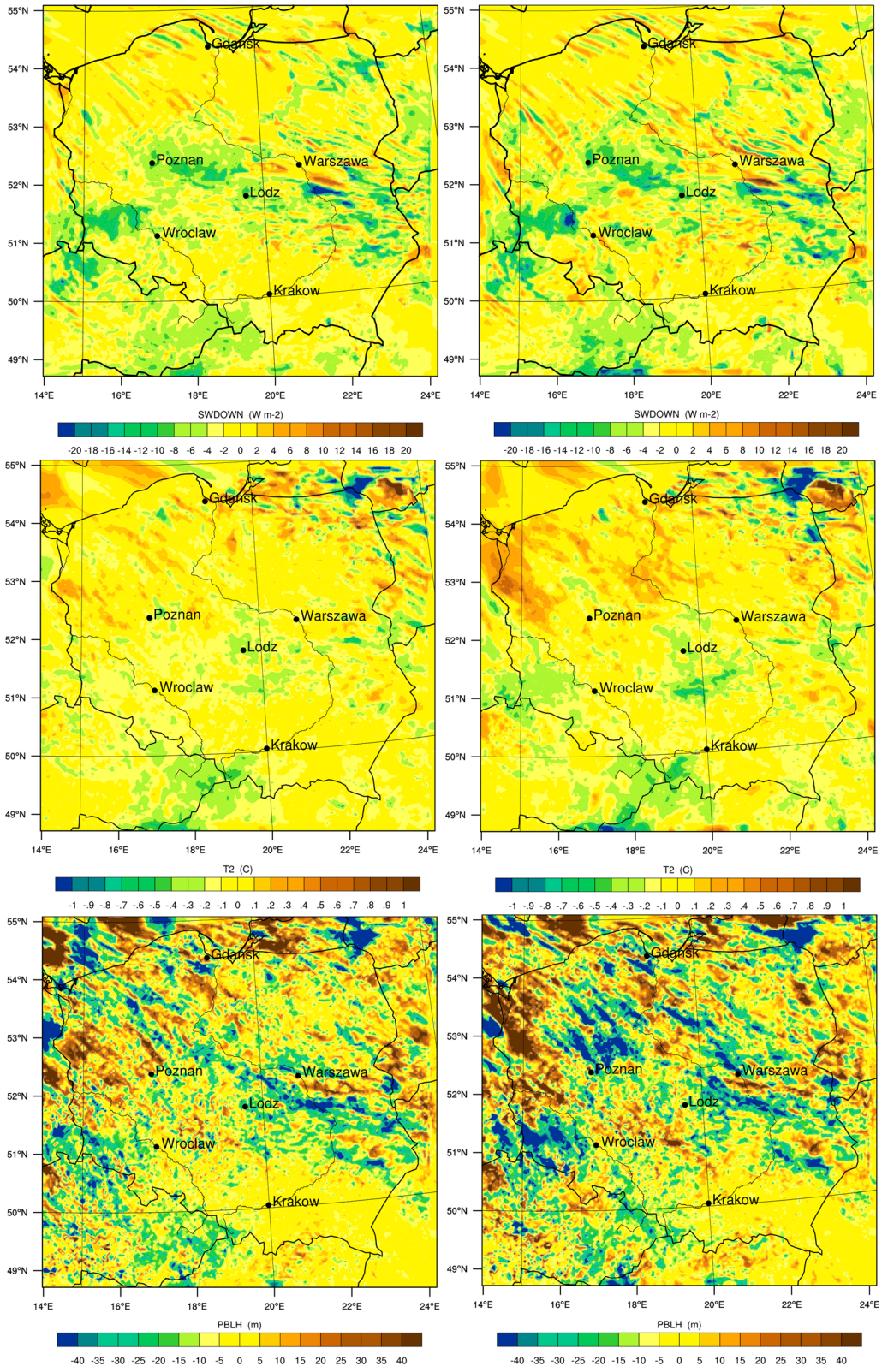
Table 4

Percentage area with differences between simulations in the following ranges given in the table

SWDOWN [W m^{-2}] (monthly mean = 53.3)								
Range	≤−10	(−10, −5>	(−5, −1>	(−1, 0>	(0, 1>	(1, 5>	(5, 10>	>10
Mon DIR	0.00	14.97	82.52	2.47	0.02	0.01	0.00	0.00
Mon INDIR	0.01	22.83	76.88	0.26	0.03	0.00	0.00	0.00
19 Jan DIR	3.87	16.28	46.21	14.98	8.59	8.43	1.40	0.25
19 Jan INDIR	5.26	16.91	40.43	12.88	9.13	12.12	2.80	0.49
29 Jan DIR	2.04	30.73	66.44	0.52	0.11	0.08	0.01	0.07
29 Jan INDIR	1.97	30.74	66.46	0.54	0.12	0.09	0.01	0.06
T2 [C] (monthly mean = −3.3)								
Range	≤−1.5	(−1.5, −1.0>	(−1.0, −0.5>	(−0.5, 0>	(0, 0.5>	(0.5, 1.0>	(1.0, 1.5>	>1.5
Mon DIR	0.00	0.00	0.06	95.70	4.24	0.00	0.00	0.00
Mon INDIR	0.00	0.00	0.57	94.71	4.71	0.00	0.00	0.00
19 Jan DIR	0.13	0.24	1.27	73.53	24.18	0.53	0.09	0.03
19 Jan INDIR	0.17	0.38	1.92	64.32	32.15	0.90	0.13	0.04
29 Jan DIR	0.81	10.30	40.89	46.21	1.77	0.01	0.01	0.00
29 Jan INDIR	1.75	10.49	39.77	36.30	1.68	0.02	0.00	0.00
PBLH [m] (monthly mean = 243.3)								
Range	<−20.0	(−20, 10>	(−10, −5>	(−5, 0>	(0, 5>	(5, 10>	(10, 20>	>20
Mon DIR	0.04	7.11	33.62	45.99	12.63	0.59	0.02	0.00
Mon INDIR	0.07	7.07	31.08	44.33	16.34	1.08	0.05	0.00
19 Jan DIR	18.00	15.68	12.17	18.05	11.24	6.93	8.15	9.77
19 Jan INDIR	21.05	14.98	10.09	14.56	10.81	7.06	9.16	12.29
29 Jan DIR	8.97	23.89	32.72	27.26	4.78	1.56	0.62	0.20
29 Jan INDIR	9.63	23.95	31.01	28.41	4.40	1.57	0.74	0.29

Mon DIR—mean monthly differences between DIR and BASE, Mon INDIR mean monthly differences between INDIR and BASE, and the same for daily differences for the 19th and 29th of January


Gdansk
Poznan
Warszawa
Lodz
Wroclaw
Krakow
55°N
54°N
53°N
52°N
51°N
50°N
49°N
14°E
16°E
18°E
20°E
22°E
24°E
SWDOWN (W m-2)
-10 -8 -6 -4 -2 0 2 4 6 8 10
T2 (C)
-1 -.9 -.8 -.7 -.6 -.5 -.4 -.3 -.2 -.1 .0 .1 .2 .3 .4 .5 .6 .7 .8 .9 1
PBLH (m)
-30 -25 -20 -15 -10 -5 0 5 10 15 20 25 30


55°N
54°N
53°N
52°N
51°N
50°N
49°N
14°E
16°E
18°E
20°E
22°E
24°E
Gdansk
Poznan
Warszawa
Lodz
Wroclaw
Krakow
SWDOWN (W m-2)
-20 -18 -16 -14 -12 -10 -8 -6 -4 -2 0 2 4 6 8 10 12 14 16 18 20
T2 (C)
-1 -.9 -.8 -.7 -.6 -.5 -.4 -.3 -.2 -.1 0 .1 .2 .3 .4 .5 .6 .7 .8 .9 1
PBLH (m)
-40 -35 -30 -25 -20 -15 -10 -5 0 5 10 15 20 25 30 35 40

◀ Figure 5
Daily mean spatial differences in SWDOWN, T2 and PBLH between DIR and BASE (*left column*) and INDIR and BASE (*right column*) for the 19nd of January 2011

R between 0.67 and 0.69. However, the largest error statistics are for the simulation with no feedback effects (BASE), and inclusion of the direct and indirect effects leads to improvements in the model performance.

For three selected sites, located in the large Polish cities (Warszawa, Poznań, Łódź), time series of modelled (BASE, DIR, INDIR) and observed concentrations and scatter plots for INDIR are presented in Fig. 7. The model generally captures the variability induced by some pollution episodes (e.g. 4–7, 28–30 of January), but in some cases underestimates their magnitude. The main reason for this is the flat annual emission profile applied for anthropogenic sources. In Poland, anthropogenic emission changes seasonally for both primary PM_{10} and their gaseous precursors, especially for SNAP sector 2 emission (residential combustion) which is largely responsible for emission of PM_{10}. For INDIR the spatial distribution of MB is presented in Fig. 8. There is a tendency towards overestimation of observed values at the stations located at the sea coast, whereas for the stations located in the central and southern Poland the model has a tendency to underestimate of PM_{10} concentration.

Mean temporal changes of PM_{10} concentrations for the total domain are presented in Fig. 9. Significantly higher concentrations for DIR and INDIR in comparison to BASE are for 27th–29th of January. This episode is also quite well reproduced in Fig. 7 which presents PM_{10} concentrations for Warszawa, Poznań and Łódź.

Table 5

Domain-wide error statistics (55 stations, January 2011) for PM_{10} concentrations for the BASE, DIR and INDIR simulations (N total number of measurements)

	BASE	DIR	INDIR
N	720	720	720
FAC2	0.89	0.89	0.89
R	0.67	0.68	0.69
MB	−9.70	−9.56	−9.54
RMSE	17.84	17.58	17.55

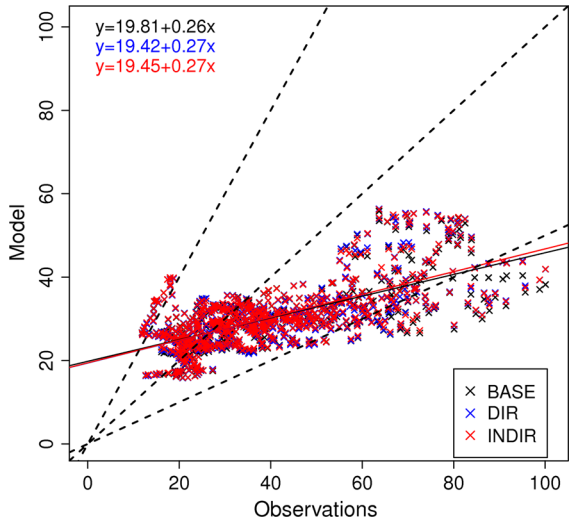


Figure 6
Scatter plot between modelled and observed PM_{10} concentrations for 55 stations for January 2011 (unit: μg m^{-3}). Different colours applied for BASE, DIR and INDIR. *P* values of fitted slopes are below 0.05

Taylor diagrams plotted according to station location (Fig. 10) show small differences with respect to the type of station. The results for background, industrial and traffic stations show very small differences. Nevertheless, the WRF-Chem results showed slightly worse results for urban stations when compared to suburban and rural. The correlation coefficients and centred RMSE for the background stations are slightly better for rural stations in comparison to suburban and urban stations.

The mean monthly modelled PM_{10} concentration for the domain of Poland (BASE simulation) in January 2011 is 26.0 μg m^{-3}. The highest concentrations concern the central part of the country and locally exceed 45.0 μg m^{-3}.

The monthly mean differences between DIR and BASE simulations range between −0.4 and 1.9 μg m^{-3} (Fig. 11). For 84 % of the area PM_{10} concentration is higher when indirect effects are present. The differences between INDIR and BASE simulations range between −0.5 and 2.0 μg m^{-3} with the spatial distribution similar to DIR.

When the direct feedback effect is included, daily differences in PM10 concentrations for the episodes of the 19th and 29th of January are between −5.0 and 13.0 μg m^{-3}. The PM_{10} concentrations are higher than for the BASE simulation for about 55 and 32 %

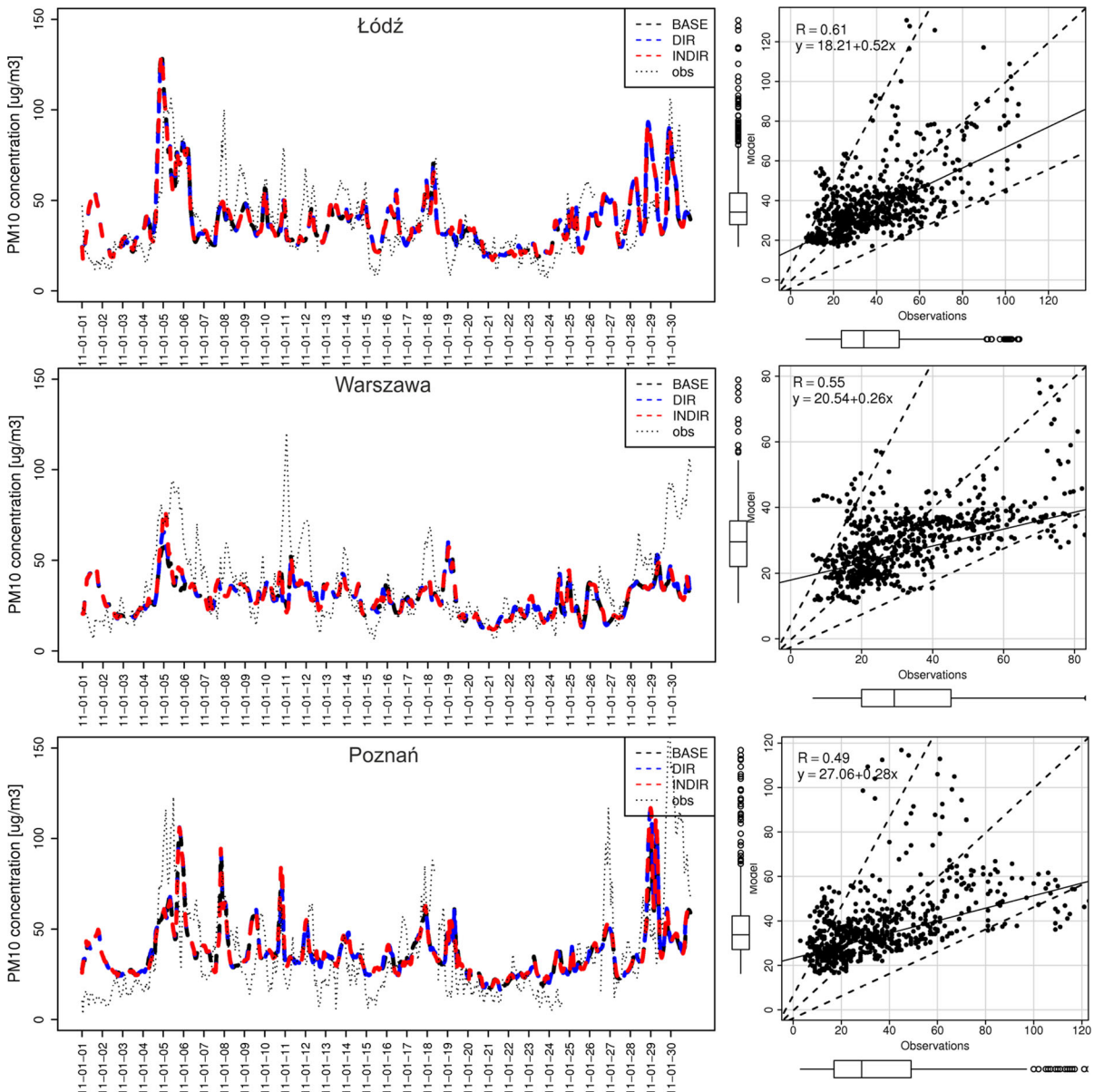


Figure 7
Time series of modelled (BASE, DIR, INDIR) and observed PM_{10} concentrations for selected stations for January 2011. *Scatter plots* of PM_{10} concentrations for the stations for INDIR simulation. *P* values of fitted slopes are below 0.05

of the domain, respectively, for the 19th and 29th of January. The highest differences are for central Poland for both 19th and 29th. In the case of the INDIR simulation the PM_{10} concentrations change in the range of −7.0 to 8.0 in comparison to DIR. The hourly variability between the DIR or INDIR and BASE simulation is higher than the daily variability. For the 19th of January at 12.00, the direct effect changes the PM_{10} concentration in the range of −13.0 and 20.0 μg m^{-3} and inclusion of the indirect effect changes the concentration from −33.0 to 28.0 μg m^{-3}, in comparison to the BASE simulation.

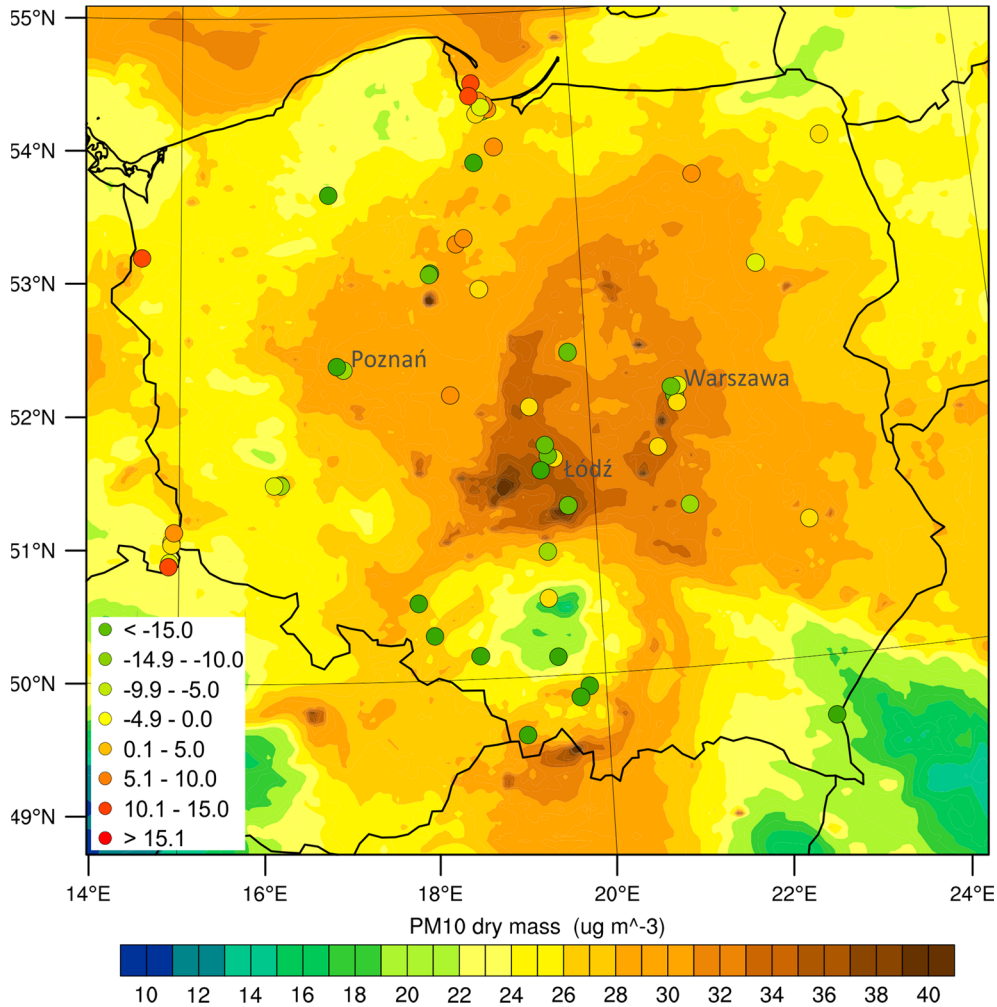


Figure 8
Monthly average PM_{10} concentrations (INDIR) and MB statistics marked by dots (MB = INDIR-observation)

4. Discussion and Conclusion

The online meteorology-chemistry model WRF-Chem has been implemented to investigate the direct and indirect feedback effects of aerosols on both meteorology and PM_{10} concentrations with the focus on the difference between specific episodes and monthly means. The simulations with high spatial resolution of 5 km × 5 km were run for Poland, for January 2011. The modelled meteorological parameters and PM_{10} concentrations have been evaluated against observations.

Aerosols affect radiation and temperature in several ways due to different radiative effects of different aerosol components (Jacobson *et al.* 1996). They can reduce incoming solar radiation via backscattering, therefore increasing the surface albedo and decreasing surface temperature. In our study a decrease in monthly mean incoming solar radiation appears for the entire area, whereas a decrease in surface air temperature is observed for about 96 % of the domain. The direct effect of aerosols on solar radiation is clearly noticeable for days with relatively high solar radiation. For these days the difference between DIR and BASE often reaches 50 W m^{-2} (e.g. between 20th and 30th January at Łódź, Fig. 3).

A higher cooling effect corresponds to the higher decrease in solar radiation. Absorption of solar radiation by black carbon and other absorbing aerosol

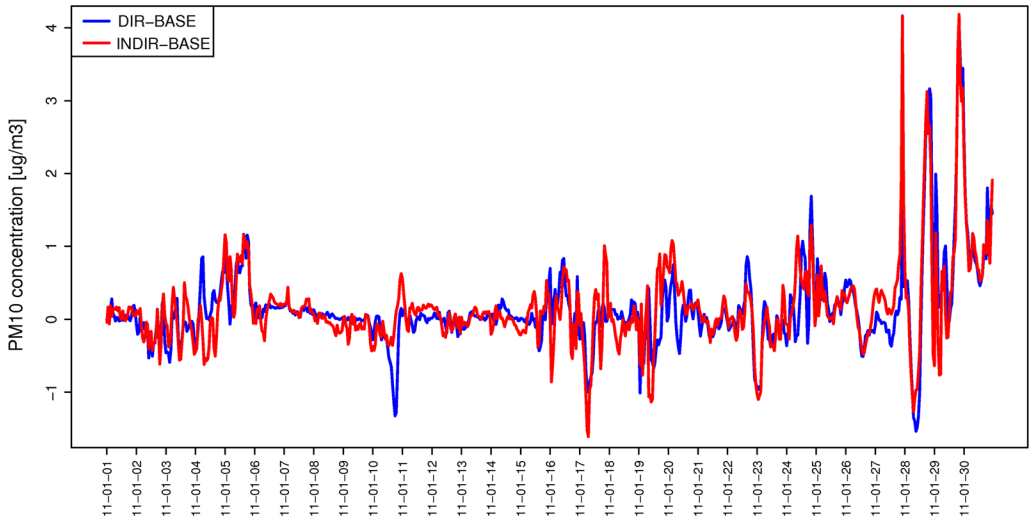


Figure 9
Time series of average differences in modelled PM_{10} concentrations between the DIR and BASE and INDIR and BASE simulations

compounds can result in regional heating of the atmosphere. Aerosols can also absorb and emit infrared radiation, also offsetting the cooling effect of backscattering during daytime. An increase in monthly mean temperature is noticed over about 4.5 % of the domain. For daily episodes on 19th and 29th January a local increase in solar radiation appears of up to 10.0 W m^{-2}. A similar effect of atmospheric heating by up to 26.0 W m^{-2} was noticed by Zhang *et al.* (2010) over the ocean and western US

Changes in air temperature in the atmosphere cause changes in monthly mean PBLH. Monthly mean PBLH changes in the range of ±5 m for 58 % of the study area. Locally the differences between DIR and BASE were higher than ±20 m. An increase appears in the northern and a decrease in the southern part of the domain. PBLH reduces because of enhanced stability as a result of the warming caused by black carbon in the PBL and the cooling at surface resulting from reduced solar radiation. Lack of this effect in the northern part of the study domain may be related to the relatively warm sea surface and local increase in air temperature. In the case of daily values the differences in PBLH for some regions reach ± 100 m. These changes may have a great impact on air pollution behaviour in the air. Reduced PBLH indicates a more stable planetary boundary layer and can thus further increase air pollution over areas where air pollution is already severe (Zhang *et al.* 2010).

Inclusion of direct feedback increases specific humidity for the western and north-western part of the domain (Fig. 3, supplementary materials). Inclusion of indirect feedback intensifies this effect for some parts of these regions. There is a strong impact of the indirect effect on specific cloud water content (QCLOUD) over the Baltic Sea and north-western and southern regions of the domain. Specific cloud water content increases notably for INDIR in comparison to the BASE and also DIR simulations and these changes are opposed to changes in Q2 (Fig. 3, supplementary material). For the region with the highest difference in QCLOUD between INDIR and BASE and relatively small difference between them, the vertical profile of QCLOUD was plotted (LOC1: 49.0°N, 20.0°E, LOC2: 52.0°N, 20.0°E, Fig. 4 in supplementary material). For the first location the vertical profile shows higher QCLOUD values for the INDIR simulation in comparison to BASE and DIR between the 1st and 5th model layers, but above these layers the results for the three simulations are the same. The highest difference between INDIR and BASE is for the 1st and 4th model layer. For the second location the QCLOUD profile is more diverse and differences appear both between the DIR and BASE and INDIR and BASE simulation. Generally, the

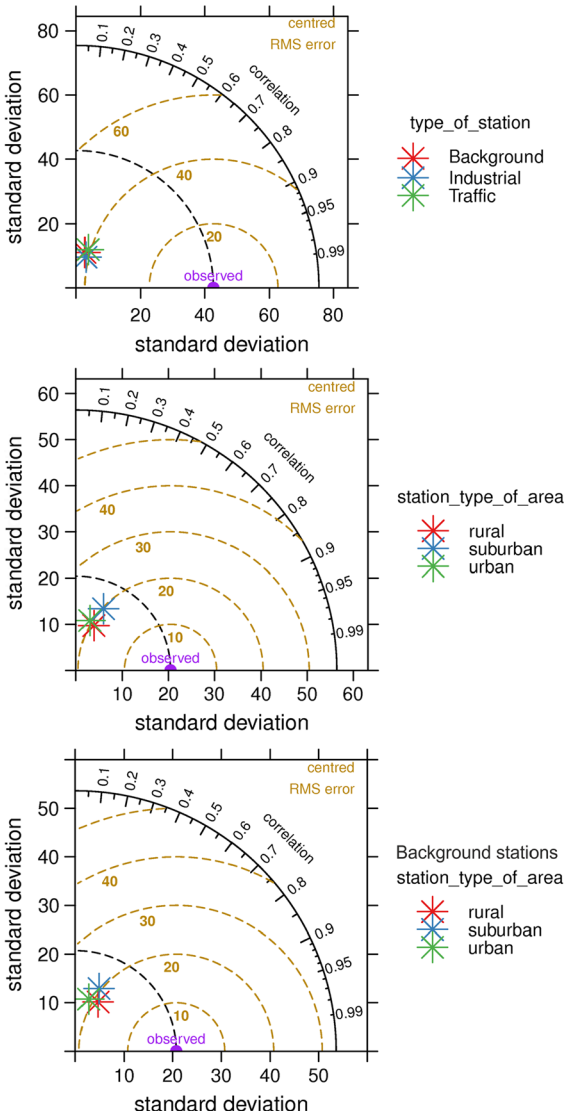


Figure 10
Taylor diagrams for PM10 concentrations for the INDIR simulation: according to type of station (background, industrial and traffic, the *upper figure*), according to station type of area (rural, suburban and rural, the *middle figure*) and using only background stations and plotted according to station area type (the *lower figure*)

highest values are for INDIR but for some areas (model layer 5) QCLOUD is the highest for BASE.

For the episodes of the 19th and 29th January, for some areas the decrease in QCLOUD is higher than 0.02 g kg^{-1} (Table 1 and Fig. 5, supplementary material). In the case of the INDIR simulation the spatial distribution of mean monthly changes is similar to DIR and ranges mainly between 0.001 and 0.300 g kg^{-1}. Considerable changes in monthly sum of rainfall for DIR simulation were found in contrast to the comparatively small changes in solar radiation for these regions. The pattern of the precipitation is not related to SWDOWN or QCLOUD.

Due to the direct effect, for 85 % of the domain, the mean monthly PM_{10} concentrations increase by up to 1.9 μg m^{-3}. A decrease of up to −0.4 μg m^{-3} was noticed over Baltic sea as well as in the western and partially also southern parts of Poland. For the INDIR simulation, generally a decrease is observed by $-0.1 \div -1.0$ μg m^{-3} in comparison to DIR but locally in central Poland an increase appears. Partially, it could be explained by liquid phase aerosols formation, as suggested in Forkel *et al.* (2012). In the case of daily values, differences in PM_{10} concentration between DIR and BASE reach 14.0 μg m^{-3} and are positive for about 80 % of the domain. Comparison of hourly PM_{10} concentrations between INDIR and BASE for the 19th and 29th of January at 12.00 gives differences of $-33.0 \div 28.0$ μg m^{-3} and $-12 \div 24$ μg m^{-3}, respectively.

The meteorological WRF model results for T2, RH2, PSFC and W10 perform well when compared with observations, with a high IOA for all parameters. Inclusion of feedback effects slightly decreases the error statistics for air temperature and relative humidity. Modelled solar radiation (SWDOWN) is in good correlation with observations, but observed values are overestimated by the model. Inclusion of feedback effects improves MB and MGE statistics. There are no large differences in mean model performance for PM_{10} concentration among the simulations. However, the worst results are obtained for the BASE simulation. The model has a tendency towards overestimation of observed PM_{10} concentrations at the sea coast station. This may be related to overestimation of sea salt aerosol emission as modelled wind speed is higher than observed values from meteorological stations in this region.

The results illustrate the potential importance of the aerosol feedback effects on modelled meteorology and PM_{10} concentrations. This influence is noticeable for mean monthly values but is evidently higher for daily and hourly episodes. This agrees well with previous studies with the COSMO-ART model on Saharan dust (Stanelle *et al.* 2010) and experiments from the EUCAARI campaign

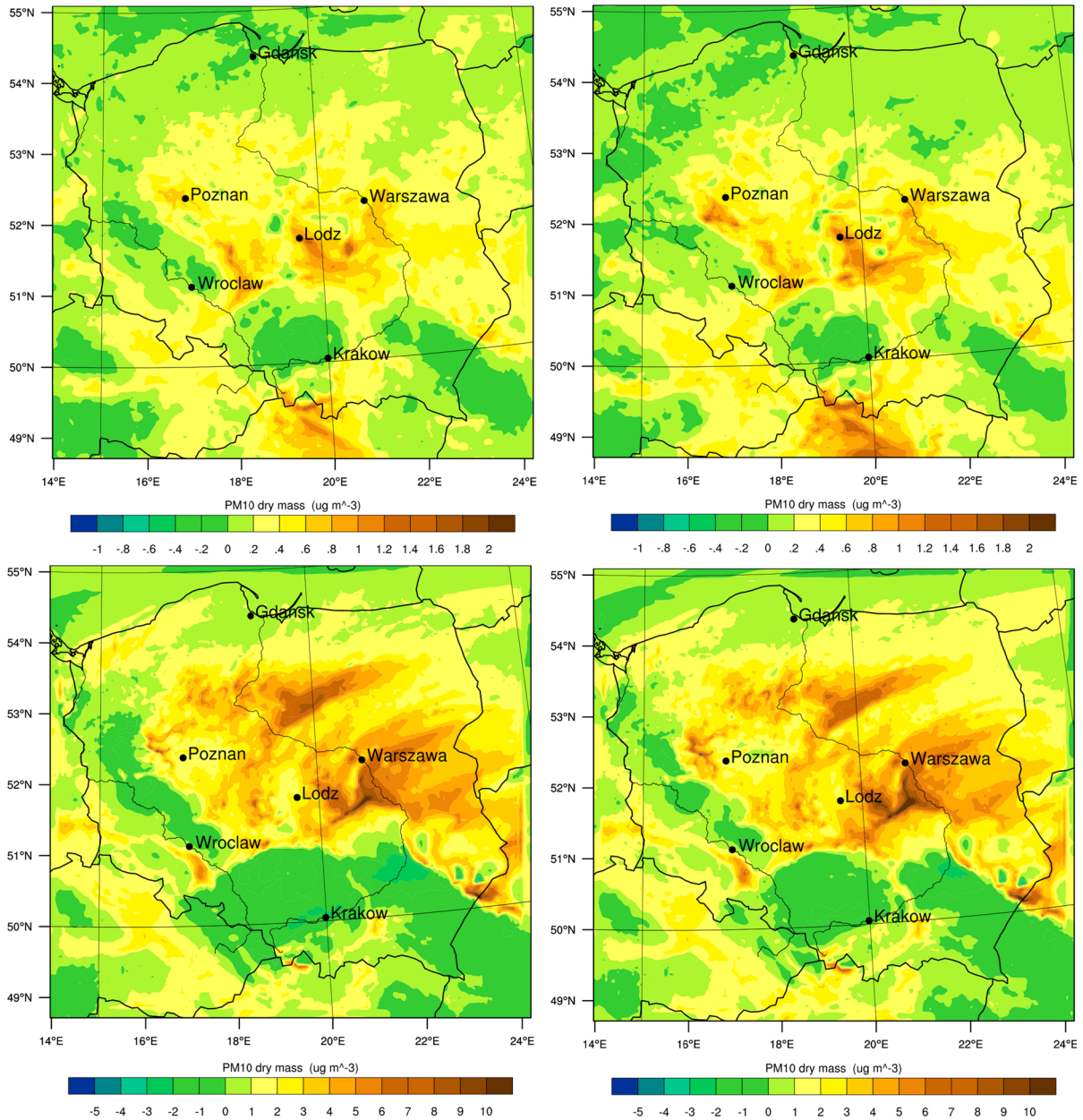


Figure 11
Mean monthly (*upper line*) and mean daily for the 29th of January spatial differences in PM_{10} concentrations between DIR and BASE (*left*) and INDIR and BASE (*right*) (different scales for monthly and daily maps)

(Athanasopoulou *et al.* 2013). We made our simulations for a winter month which due to low temperatures and limited sun-shine has limited emissions from nature. A summer simulation should, therefore, provide a much higher impact from feedback effects. This suggests that for studies in high temporal resolution the online models are necessary to describe the processes and feedback effects correctly to obtain the most reliable results. For long-term studies the offline models in most cases meet the requirements as the overall feedback effect is reduced over longer periods. This study has been based on the winter period, which is characterised in Poland by high anthropogenic emissions of particulate matter

and severe meteorological conditions. However, it was noticed that the highest feedback effects are not strictly related with the highest particulate matter concentrations. We suggest to carry out a similar study for this region for the summer period, which is characterised by higher solar radiation, high BVOC emissions from nature and more dynamic PBL, or during episodes with substantial transport of particles due to Saharan dust.

Acknowledgments

The authors acknowledge the Aerosol Research Network Poland-AOD and the Institute of Geophysics, University of Warsaw for the provision of radiation data, the Institute of Meteorology and Water management for the provision of measurements of: air temperature, relative humidity, surface pressure and wind speed and the Chief Inspectorate of Environmental Protection in Poland for PM_{10} measurements. This study has received support from the Polish National Science Centre through project no. UMO-2013/09/B/ST10/00594.

References

Ackermann, I.J., Hass, H., Memmesheimer, M., Ebel, A., Binkowski, F.S., Shankar, U., (1998). *Modal aerosol dynamics model for Europe*. Atmos. Environ. *32*, 2981–2999.

Andreae, M.O., Jones, C.D., Cox, P.M., (2005. *Strong present-day aerosol cooling implies a hot future*. Nature *435*, 1187–90.

Athanasopoulou, E., Vogel, H., Vogel, B., Tsimpidi, A. P., Pandis, S. N., Knote, C., Fountoukis, C., (2013). *Modeling the meteorological and chemical effects of secondary organic aerosols during an EUCAARI campaign*, Atmos. Chem. Phys., *13*(2), 625–645, doi:10.5194/acp-13-625-2013.

Baklanov, A., Schlünzen, K.., Suppan, P., Baldasano, J., Brunner, D., Aksoyoglu, S., Carmichael, G., Douros, J., Flemming, J., Forkel, R., Galmarini, S., Gauss, M., Grell, G., Hirtl, M., Joffre, S., Jorba, O., Kaas, E., Kaasik, M., Kallos, G., Kong, X., Korsholm, U., Kurganskiy, a., Kushta, J., Lohmann, U., Mahura, a., Manders-Groot, a., Maurizi, a., Moussiopoulos, N., Rao, S.T., Savage, N., Seigneur, C., Sokhi, R.S., Solazzo, E., Solomos, S., Sørensen, B., Tsegas, G., Vignati, E., Vogel, B., Zhang, Y., (2014. *Online coupled regional meteorology chemistry models in Europe: current status and prospects*. Atmos. Chem. Phys. *14*, 317–398.

Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hansen, J.E., Hofmann, D.J., (1992). *Climate forcing by anthropogenic aerosols*. Science *255*, 423–30.

Chen, F., Dudhia, J., (2001). *Coupling an Advanced Land Surface–Hydrology Model with the Penn State–NCAR MM5 Modeling System. Part I: Model Implementation and Sensitivity*. Mon. Weather Rev. *129*, 569–585.

Fast, J.D., Gustafson, W.I., Easter, R.C., Zaveri, R.A., Barnard, J.C., Chapman, E.G., Grell, G.A., Peckham, S.E., (2006). *Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model*. J. Geophys. Res. *111*, D21305.

Forkel, R., Werhahn, J., Hansen, A.B., McKeen, S., Peckham, S., Grell, G., Suppan, P., (2012). *Effect of aerosol-radiation feedback on regional air quality – A case study with WRF/Chem*. Atmos. Environ. *53*, 202–211.

Grell, G. A., Peckham, S.E., Schmitz, R., McKeen, S. a., Frost, G., Skamarock, W.C., Eder, B., (2005). *Fully coupled "online" chemistry within the WRF model*. Atmos. Environ. *39*, 6957–6975. doi:10.1016/j.atmosenv.2005.04.027.

Grell, G.A., (2002). *A generalized approach to parameterizing convection combining ensemble and data assimilation techniques*. Geophys. Res. Lett. *29*, 1693.

Hong, S.Y., Noh, Y., Dudhia, J., (2006). *A New Vertical Diffusion Package with an Explicit Treatment of Entrainment Processes*. Mon. Weather Rev. *134*, 2318–2341.

Iacono, M.J., Delamere, J.S., Mlawer, E.J., Shephard, M.W., Clough, S.A., Collins, W.D., (2008). *Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models*. J. Geophys. Res. *113*, D13103.

Jacobson, M.Z., Lu, R., Turco, R.P., Toon, O., (1996). *Development and application of a new air pollution model system e Part I: Gas-phase simulations*. Atmos. Env *30*, 1939–1963.

Jacobson, M., Z., (2002). Atmospheric Pollution History, Science, and Regulation, Cambridge University Press.

Kaminski, J., Neary, L., Lupu, A., McConnell, J., Struzewska, J., Zdunek, M., Lobocki, L., (2007). High Resolution Air Quality Simulations with MC2-AQ and GEM-AQ, in: Borrego, C., Norman, A.-L. (Eds.), Air Pollution Modeling and Its Application XVII SE - 86. Springer US, pp. 714–720.

Kuenen, J.J.P., Visschedijk, a. J.H., Jozwicka, M., Denier van der Gon, H. A. C., (2014). *TNO-MACC_II emission inventory: a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling*. Atmos. Chem. Phys. Discuss. *14*, 5837–5869.

Kryza, M., Wałaszek, K., Ojrzyńska, H., Szymanowski, M., Werner, M. and Dore, A. J., (2015). High resolution dynamical downscaling of ERA-Interim using the WRF regional climate model (Part 1) – model configuration and statistical evaluation for the 1981-2010 period, Pure Appl. Geophys., in review.

Lin, Y.L., Farley, R.D., Orville, H.D., (1983). *Bulk Parameterization of the Snow Field in a Cloud Model*. J. Clim. Appl. Meteorol. *22*, 1065–1092.

Liu, S.C., McKeen, S.A., Hsie, E.-Y., Lin, X., Kelly, K.K., Bradshaw, J.D., Sandholm, S.T., Browell, E. V., Gregory, G.L., Sachse, G.W., Bandy, A.R., Thornton, D.C., Blake, D.R., Rowland, F.S., Newell, R., Heikes, B.G., Singh, H., Talbot, R.W., (1996). *Model study of tropospheric trace species distributions during PEM-West* A. J. Geophys. Res. *101*, 2073.

Lynn, B., Khain, A., Rosenfeld, D., Woodley, W.L., (2007). *Effects of aerosols on precipitation from orographic clouds*. J. Geophys. Res. *112*, D10225.

Mercado, L.M., Bellouin, N., Sitch, S., Boucher, O., Huntingford, C., Wild, M., Cox, P.M., (2009). *Impact of changes in diffuse radiation on the global land carbon sink*. Nature *458*, 1014–7.

Moran, M.D., Ménard, S., Talbot, D., Huang, P., Makar, P.A., W., G., Landry, H., Gravel, S., Gong, S., Crevier, L.-P., K.A., Sassi, M., (2010). Air Pollution Modeling and Its Application XX, in: Steyn, D.G., Rao, S. (Eds.), Air Pollution Modelling and Its Application XX. Springer Science & Business Media, pp. 289–292.

Perez, L., Tobias, A., Querol, X., Künzli, N., Pey, J., Alastuey, A., Viana, M., Valero, N., González-Cabré, M., Sunyer, J., (2008). *Coarse particles from Saharan dust and daily mortality*. Epidemiology *19*, 800–7.

Pope, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K., Thurston, G.D., (2002). *Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution*. JAMA *287*, 1132–41.

Rosenfeld, D., Lohmann, U., Raga, G.B., O'Dowd, C.D., Kulmala, M., Fuzzi, S., Reissell, A., Andreae, M.O., (2008). *Flood or drought: how do aerosols affect precipitation?* Science *321*, 1309–13.

Schell, B., Ackermann, I.J., Hass, H., Binkowski, F.S., Ebel, A., (2001). *Modeling the formation of secondary organic aerosol within a comprehensive air quality model system*. J. Geophys. Res. Atmos. *106*, 28275–28293.

Stanelle, T., Vogel, B., Vogel, H., Bäumer, D., Kottmeier, C., (2010). Feedback between dust particles and atmospheric processes over West Africa during dust episodes in March 2006 and June 2007, Atmos. Chem. Phys., 10(22), 10771–10788, doi:10.5194/acp-10-10771-2010.

Stockwell, W.R., Middleton, P., Chang, J.S., Tang, X., (1990). The second generation regional acid deposition model chemical mechanism for regional air quality modeling. J. Geophys. Res. 95, 16343.

Tuccella, P., Curci, G., Visconti, G., Bessagnet, B., Menut, L., Park, R.J., (2012). *Modeling of gas and aerosol with WRF/Chem over Europe: Evaluation and sensitivity study*. J. Geophys. Res. *117*, D03303.

Wilton, D.J., Hewitt, C.N., Beerling, D.J., (2011). *Simulated effects of changes in direct and diffuse radiation on canopy scale isoprene emissions from vegetation following volcanic eruptions*. Atmos. Chem. Phys. *11*, 11723–11731.

Zhang, Y., 2008. *Online coupled meteorology and chemistry models: history, current status, and outlook*. Atmos. Chem. Phys. Discuss. *8*, 1833–1912.

Zhang, Y., Wen, X.Y., Jang, C.J., (2010). *Simulating chemistry–aerosol–cloud–radiation–climate feedbacks over the continental U.S. using the online-coupled Weather Research Forecasting Model with chemistry (WRF/Chem)*. Atmos. Environ. *44*, 3568–3582.

(Received November 26, 2014, accepted March 3, 2016)