Environmental Impact Statement to manuscript "A shift in emission time profiles of fossil fuel combustion due to energy transitions impacts source receptor matrices for air quality" by Carlijn Hendriks, Jeroen Kuenen, Richard Kranenburg, Yvonne Scholz and Martijn Schaap

The work presented here shows that the time (of day and year) at which emissions of air pollutants occur impacts ambient pollutant concentrations, and that a shift in emission timing changes source receptor relationships. This finding is relevant when the impact of energy transitions on air quality is assessed since the deployment of renewable electricity technologies not only lowers emissions from fossil-fuel power plants but also makes the timing of these emissions more dependent on synoptic conditions, which might limit the gain in air quality because of the emission reduction. Therefore, to accurately assess the impact of energy transitions on air quality source receptor relations for each scenario should be established before calculating the impacts on ecosystems and human health.

A shift in emission time profiles of fossil fuel combustion due to energy transitions impacts source receptor matrices for air quality

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Effective air pollution and short-lived climate forcer mitigation strategies can only be designed when the effect of emission reductions on pollutant concentrations and health and ecosystem impacts are quantified. Within integrated assessment modeling source-receptor relationships (SRRs) based on chemistry transport modeling are used to this end. Currently, these SRRs are made using invariant emission time profiles. The LOTOS-EUROS model equipped with a source attribution module was used to test this assumption for renewable energy scenarios. Renewable energy availability and thereby fossil fuel back up are strongly dependent on meteorological conditions. We have used the spatially and temporally explicit energy model REMix to derive time profiles for backup power generation. These time profiles were used in LOTOS-EUROS to investigate the effect of emission timing on air pollutant concentrations and SRRs. It is found that the effectiveness of emission reduction in the power sector is significantly lower when accounting for the shift in the way emissions are divided over the year and the correlation of emissions with synoptic situations. The source receptor relationships also changed significantly. This effect was found for both primary and secondary pollutants. Our results indicate that emission timing deserves explicit attention when assessing the impacts of system changes on air quality and climate forcing from short lived substances.

1. Introduction

Global energy consumption has grown considerably over the last decades and is anticipated to grow further in the future[1]. To date, a large share of the energy used originates from fossil fuels. To reduce the impact of energy use on climate, the European Commission has set goals to increase the share of renewable energies in Europe to 20% by 2020[2]. One of the major pathways leading to a sustainable energy system is electrification of transport and the building sector[3], in combination with using renewable energy sources for the electricity generation sector. According to the Roadmap towards a low carbon economy in 2050 in Europe, greenhouse gas emissions from the power sector should be reduced by 54 – 68% in 2030 and 93 – 99% in 2050[4]. A major role in a sustainable power sector is often attributed to wind and especially solar (photovoltaic, PV) energy, since these are available in abundance throughout Europe and beyond[1,3]. Bioenergy is also anticipated to become more important, but will mainly be used as direct fuel and not for electricity production[5].

Solar and wind based electricity systems are intermittent power sources, i.e., the electricity production depends on weather conditions and availability of sunlight. Consequently, electricity demand cannot be met at each hour of the day and night by PV and wind power alone. This could be accounted for by storing energy when it is abundantly available but this is relatively expensive and difficult to achieve[6,7,8]. As long as energy storage is not a viable option on the scale required, there is a need for back-up electricity generation capacity that can be switched on and off quickly, to be used when the supply of electricity from renewable technologies is insufficient. In the coming decades, the back-up capacity most likely consists of fossil fuel (especially natural gas) fired plants. However, considering that the price of coal is much lower than for natural gas, coal fired power plants may also be used[9]. Hence, a solid environmental impact assessment for fossil fuel combustion remains necessary in the future.

Currently, power generation is an important contributor to atmospheric concentrations of air pollutants like sulfur dioxide (SO_2), nitrogen dioxide (NO_2) and particulate matter (PM)[10,11]. Exposure to these pollutants is associated with adverse health effects[12,3] and loss of biodiversity[13]. Furthermore, these pollutants contribute to climate forcing through aerosols and ozone[14,15]. As pollutant emissions from fossil fuel fired power plants will be reduced dramatically and emissions from renewable electricity generation are much smaller, a transition to renewable energy will have a significant impact on air quality[16]. Given the intermittent nature of renewables, there will also be a significant change in the temporal variability of the emissions. At the moment, as most power plants are fossil fuel based, the highest emissions from power plants occur when the demand for electricity is highest. When renewables provide a large share of electricity demand, the highest emissions will occur when the gap between the renewable electricity production and electricity demand is largest. Air pollutant concentrations and fate are dependent on meteorological conditions and chemical regime and are thus impacted by seasonal and diurnal emission timing patters[17,18,19]. A shift in the temporal variability of the emissions could therefore impact the relation between an emission from a certain source and its impact on air pollutant concentrations in a certain receptor region, also called source receptor relations (SRRs).

Source receptor relations are commonly used in integrated assessment models to assess the impact of emission reduction measures and design cost effective mitigation strategies[20,21]. These models are widely applied for policy support and political negotiations are informed by the outcome of integrated assessment modeling studies. In these models, the SRRs are assumed to be linear and constant, enabling fast calculations of the expected effect of mitigation measures. Currently, SSRs are calculated by reducing one by one the pollutant emission total (by a fixed relative amount) from each country in Europe[21,22]. Except for the emission total all model parameters, including temporal emission patterns, are kept constant. Currently, integrated assessment models are extended to be able to assess co-benefits between air pollution and climate policies[20,23]. Hence, for the application to energy transition scenarios the sensitivity of the SRRs to shifts in emission time profiles needs to be known.

In this study, we explore the impact of changing time profiles of emissions from the power sector on source receptor relations. We developed two simple renewable energy scenarios by assuming a certain share of wind and PV power in the electricity mix (Section 2). The corresponding emission time profiles were developed by hourly matching of electricity production and consumption (section 2). The air quality impacts of these scenarios and the impacts on SRRs are assessed using the Chemistry Transport Model (CTM) LOTOS-EUROS (Section 3). Results are provided in section 4 and discussed in section 5.

2. Scenario definition

In this study, four emission scenarios for Europe (in this study taken as the European Union plus Norway, Croatia, Turkey and Switzerland) were defined to investigate the effect of a shift in temporal variability associated with a high deployment of renewable electricity on air quality. For the baseline scenario, the current electricity mix is used, consisting of fossil fuels (55% of the electricity generated in Europe), nuclear power (27%) hydroelectric power (including pumped storage) (16%), wind (2%) and other sources, including solar energy (together adding up to 0.3%).[24] Between countries, large differences in the electricity mix exist. For example, France and Norway have much higher shares than average of nuclear and hydro power, respectively. In the scenarios with high renewable electricity production, the share of renewable electricity production (i.e. PV and wind power) is increased, replacing fossil-fuel based electricity.

To keep the scenarios as simple as possible, storage and trade of electricity are not included in our scenarios. This means that for each hour and each country the electricity load should equal the sum of the electricity generation from all sources:

$$\mathrm{U_{total}(x,t) = U_{nuclear,hydro}(x) + U_{PV}(x,t) + U_{wind(onshore)}(x,t) + U_{wind(offshore)}(x,t) + U_{fossil}(x,t))}$$

Here, $U_{total}(x,t)$ is the electricity demand for country *x* at hour *t*. $U_{nuclear,hydro}(x)$ is the contribution of hydroelectric and nuclear power. These sources are assumed to generate a constant power output each hour of the year, making $U_{nuclear,hydro}$ time-independent. $U_{PV}(x,t)$, $U_{wind(onshore)}(x,t)$ and $U_{wind(offshore)}(x,t)$ represent the electricity generated by the three renewable sources considered in this study, and $U_{fossil}(x,t)$ is the remaining fossil fuel needed to fulfill the demand.

The production of renewable electricity for each hour (is defined using the following equation:

$$\mathrm{U_{renewable,total}}(x) = \sum_{Renewable\ Types} \left(\alpha_{Ren}(x) \times \sum_{t=1}^{t=8760} P_{Ren}(x,t) \right)$$

The potentials $P_{Ren}(x,t)$ represent the electricity generation from a renewable source that would be possible for country *x* at hour *t* if the maximum capacity for that source in country *x* would be installed, whereas α represents the fraction of the maximum capacity that is installed in country *x* in a scenario. Hourly renewable electricity generation potentials $P_{ren}(x,t)$ were calculated using the REMix (Renewable Energy Mix for Sustainable Electricity Supply) model[25]. REMix is an energy system model that calculates the hourly availability of renewable electricity based on meteorological conditions. The energy system model can also dimension power supply systems with high shares of renewable energy and calculate the least cost operation of the system components.

For the baseline scenario, $\alpha_{Ren}(x)$ are chosen such that over the whole year, the current contributions of PV and wind to the electricity mix of each country are obtained. In the first renewable energy scenario, hereafter referred to as the *50/50 scenario*, $\alpha_{Ren}(x)$ are chosen such that over the whole year, the contributions of PV and wind to the electricity mix are approximately equal, together totalling 30% of the electricity demand. In the second scenario, the *high wind scenario*, $\alpha_{Wind}(x)$ is chosen such that wind energy produces 30% of the electricity demand. Where the 30% is not reached, PV power is used to fill the gap.

The determination of α for all renewable technologies was done iteratively, starting by choosing α such that $U_{renewable,total}(x)$ meets the requested share of the total electricity generation. However, for some *x, t* combinations there is overproduction of electricity from renewables. Since no storage or trading is assumed, this electricity is "lost" and the parameter α needs to be increased to reach the envisaged contribution of renewables. This iterative procedure has been repeated until no further improvement was found. However, the 30% contribution of renewables is not reached Europe-wide. This is due to the fact that in some countries (e.g. France and Norway) the power production from nuclear and/or hydro installations is so large that the share of of PV and wind together cannot reach the 30% by replacing only fossil fuel based energy. Therefore, in both scenarios with high renewable deployment, the renewable share in the whole region is around 25%. The share of fossil fuels in the electricity mix is 57% in the baseline scenario and 34% for the 50/50 and high wind scenarios.
The assumption that hydroelectric power generation is constant throughout the year is an over-simplification as well: in reality it can be varied according to the demand. Therefore, the scenarios developed in this study should not be seen as realistic, but merely as a means to explore the impact of a shift in time profiles of emissions from power plants in Europe on air quality.

The annual total emissions for all sectors are taken from the TNO-MACC database[26]. In all scenario runs except the baseline, the emissions of the power sector are reduced by the percentage of fossil fuels replaced by renewables. Therefore, in the scenarios assuming a high deployment of renewable electricity, the annual emissions from the European power sector are effectively reduced by 40%. In this study, we have assumed that the emissions are reduced equally across all power plants. Also, it has been assumed that the shares of each fuel in the fossil fuel generated electricity remains constant. In the real world, some power plants would be shut down completely and others would remain fully operational and fuel shift is possible, but including this is beyond the scope of this study.
For all scenarios including the baseline, the annual total emissions from the power sector for each country were divided over the year assuming a linear relation to the fossil fuel based electricity generated:

$$E_i(x,t) = \frac{U_{fossil}(x,t)}{U_{fossil,total}(x)} \cdot E_i(x)_{total}$$

Here, $E_i(x,t)$ is the emission of substance *i* in country *x* at hour *t* and $E_i(x)_{total}$ is the annual emission of that substance in that country.
Additional to the baseline, 50/50 and high wind scenarios, a control scenario was defined to be able to distinguish the impact of the emission reduction and of the change in timing. This 'low emission' scenario consists of the emission totals of the '50/50' scenario and the time profiles of the baseline scenario.
The distribution of the fossil-based electricity varies considerably between the scenarios (Table 1). In the equal PV/wind scenario, the relative difference between summer and winter becomes larger due to the abundant availability of PV power in the summer months. The high wind scenario shows more fluctuations throughout the year because high wind speed conditions come in episodes. For the equal PV/wind scenario, these fluctuations are partly subdued by using two renewable sources, each with its own favorable weather conditions.

Table 1: Energy mix in several countries (Czech Republic (CZE), Germany (DEU), France (FRA), the Netherlands (NLD)) for the energy scenarios used in this research. Shares of fossil, solar, wind and other

electricity sources for the whole year, in summer (june/july/august) and winter (nov/dec/jan).

		Whole Year				Summer				Winter			
		Fossil	PV	Wind	Other	Fossil	PV	Wind	Other	Fossil	PV	Wind	Other
Reference	CZE	64.7%	0.0%	0.1%	35.3%	59.4%	0.0%	0.0%	40.5%	69.3%	0.0%	0.1%	30.6%
50/50	CZE	31.5%	20.4%	12.8%	35.3%	21.8%	29.1%	8.5%	40.5%	43.3%	11.9%	14.1%	30.6%
High wind	CZE	37.5%	0.0%	27.2%	35.3%	39.2%	0.0%	20.3%	40.5%	40.6%	0.0%	28.8%	30.6%
Reference	DEU	60.4%	3.4%	4.8%	31.4%	58.1%	5.2%	3.0%	33.8%	64.2%	1.7%	5.0%	29.2%
50/50	DEU	39.1%	14.8%	14.8%	31.4%	35.0%	22.2%	9.0%	33.8%	47.8%	7.2%	15.7%	29.2%
High wind	DEU	36.2%	3.4%	29.1%	31.4%	42.8%	5.2%	18.3%	33.8%	38.4%	1.6%	30.8%	29.2%
Reference	FRA	14.1%	0.0%	0.7%	85.1%	3.1%	0.0%	0.2%	96.6%	27.7%	0.0%	0.9%	71.3%
50/50	FRA	4.1%	5.1%	5.7%	85.1%	0.0%	2.5%	0.8%	96.6%	11.1%	7.3%	10.2%	71.3%
High wind	FRA	3.8%	0.0%	11.0%	85.1%	0.8%	0.0%	2.6%	96.6%	9.9%	0.0%	18.8%	71.3%
Reference	NLD	92.8%	0.7%	2.8%	3.8%	93.3%	1.0%	1.7%	3.9%	92.9%	0.3%	3.0%	3.7%
50/50	NLD	70.0%	11.2%	15.0%	3.8%	70.2%	16.5%	9.4%	3.9%	75.0%	5.0%	16.2%	3.7%
High wind	NLD	65.6%	0.7%	29.9%	3.8%	76.2%	1.0%	18.8%	3.9%	63.6%	0.3%	32.3%	3.7%

3. Model description

The scenarios described above were used as input to the chemistry transport model LOTOS-EUROS[27] version 1.8 to calculate the effects of a high deployment of solar and wind energy on air pollutant concentrations. Four simulations (one for each scenario) were carried out for the European domain (13° East – 35° West, 35 – 70° South). The model uses a normal longitude–latitude projection at a standard grid resolution of 0.50° × 0.25° (longitude x latitude). The model top is placed at 3.5 km above sea level and consists of three dynamical layers: a mixing layer and two reservoir layers on top. The height of the mixing layer at each time and position is extracted from ECMWF meteorological data used to drive the model. The height of the reservoir layers is set to the difference between ceiling (3.5 km) and mixing layer height. Both layers are equally thick with a minimum of 500 m. If the mixing layer is near or above 3500 m high, the top of the model exceeds 3500 m. A surface layer with a fixed depth of 25 m is included in the model to monitor ground-level concentrations.

Advection in all directions is described using the monotonic advection scheme developed by Walcek[28]. Gas phase chemistry is handled with the TNO CBM-IV scheme[29], which is based on the scheme by Whitten et al.[30]. Hydrolysis of N_2O_5 is described following Schaap et al.[31]. Aerosol chemistry is represented with ISORROPIA2[32]. The pH dependent cloud chemistry scheme follows Banzhaf et al.[33]. Formation of course-mode nitrate is included in a dynamical approach[34]. Dry deposition for gases is modeled using the DEPAC3.11 module, which includes canopy compensation points for ammonia deposition[34, 35]. Deposition of particles is represented following Zhang et al.[36]. Stomatal resistance is described by the parameterization of Emberson et al.[37,38] and the aerodynamic resistance is calculated for all land use types separately. Wet deposition is handled using simple scavenging coefficients for gases[39] and particles[40]. The CORINE land use dataset[41] combined with the distributions of 115 tree species over Europe[42] are used to calculate biogenic VOC emissions following Schaap et al.[29], which is comparable to the approach of Steinbrecher[43]. Emissions from wild fires and boundary conditions are taken from the global MACC service[44]. Anthropogenic emissions are taken from the TNO-MACC database[26]. The treatment of the power sector is discussed in detail in the previous section. The temporal variation of the emissions from other sectors is represented by monthly, daily and hourly time factors for each source category[45]. The emission height distribution for all source sectors follows the Eurodelta approach[46]. For all sectors, elemental carbon (EC) is calculated as a fraction of the primary particulate matter (PPM) emission. This fraction is country and sector dependent.

Previous versions of the model have been used for the assessment of (particulate) air pollution[29 31,39,47,48,49]. The model has participated frequently in international model comparisons aimed at ozone[50,51], PM[52,53] and source receptor matrices[54]. For a detailed description of the model we refer to Schaap et al.[27],Kranenburg et al.[55], Wichink Kruit et al.[34] and abovementioned studies.

Source apportionment module

A source apportionment module for LOTOS-EUROS was developed to be able to track the origin of NOx, SO2 and PM10 and its components[55]. This module uses a labelling approach similar to the approach taken by Wagstrom et al.[56], tracking the source contribution of a set of sources through the model system. The emissions can be categorized and labelled in several source categories (e.g. countries, sector, fuel type) before the model is executed. The total concentration of each substance in each grid cell is modelled as usual. Additionally, the fractional contribution of each label to the total concentration of every species is calculated. During or after each process, the new fractional contribution of each label is defined by calculating a weighted average of the fractions before the process and the concentration change during the process. For details and validation of this source apportionment module we refer to Kranenburg et al.[55]. In this study, emissions from power plants were given a separate label to distinguish them from emissions from other sectors. Ten countries across Europe were selected and labeled separately in order to calculate the effect from emissions from these countries on the whole domain. This resulted in 24 labels, including labels for natural emissions and for influx from outside the model domain. The labels are listed in Table 2.

Table 2: Overview of the labels used in all scenario runs.

Label	Country	Emission source
1	Spain	Power plants
2	Great Britain	Power plants
3	Germany	Power plants
4	France	Power plants
5	Italy	Power plants
6	Poland	Power plants
7	Czech Republic	Power plants
8	Belgium and Luxembourg	Power plants
9	The Netherlands	Power plants
10	Other countries	Power plants
11	Spain	Other sectors
12	Great Britain	Other sectors
13	Germany	Other sectors
14	France	Other sectors
15	Italy	Other sectors
16	Poland	Other sectors
17	Czech Republic	Other sectors
18	Belgium and Luxembourg	Other sectors
19	The Netherlands	Other sectors
20	Other countries	Other sectors
21	-	Natural sources
22, 23, 24	-	Sources outside model domain

4. Results

To investigate the impacts of a change in the electricity generation system on air quality, we focus on sulfur dioxide (SO_2), particulate sulfate (SO_4), nitrogen dioxide (NO_2), nitrate (NO_3), total particulate matter (PM_{10}) and elemental carbon (EC). All these pollutants (NO_x, SO_2, EC, primary particulate matter (PPM), some SO_4) and the precursors of secondary PM (SO_4 and NO_3) are emitted during combustion processes in power plants. While NO_x emissions are almost independent of fuel type, SO_2 and SO_4 are emitted mostly during coal combustion. Emissions of PM (including EC) and PM precursors also differ with fuel and technology.

First, we validate model performance for these substances, after which the concentrations and contributions from the power sector for each scenario are presented.

Validation

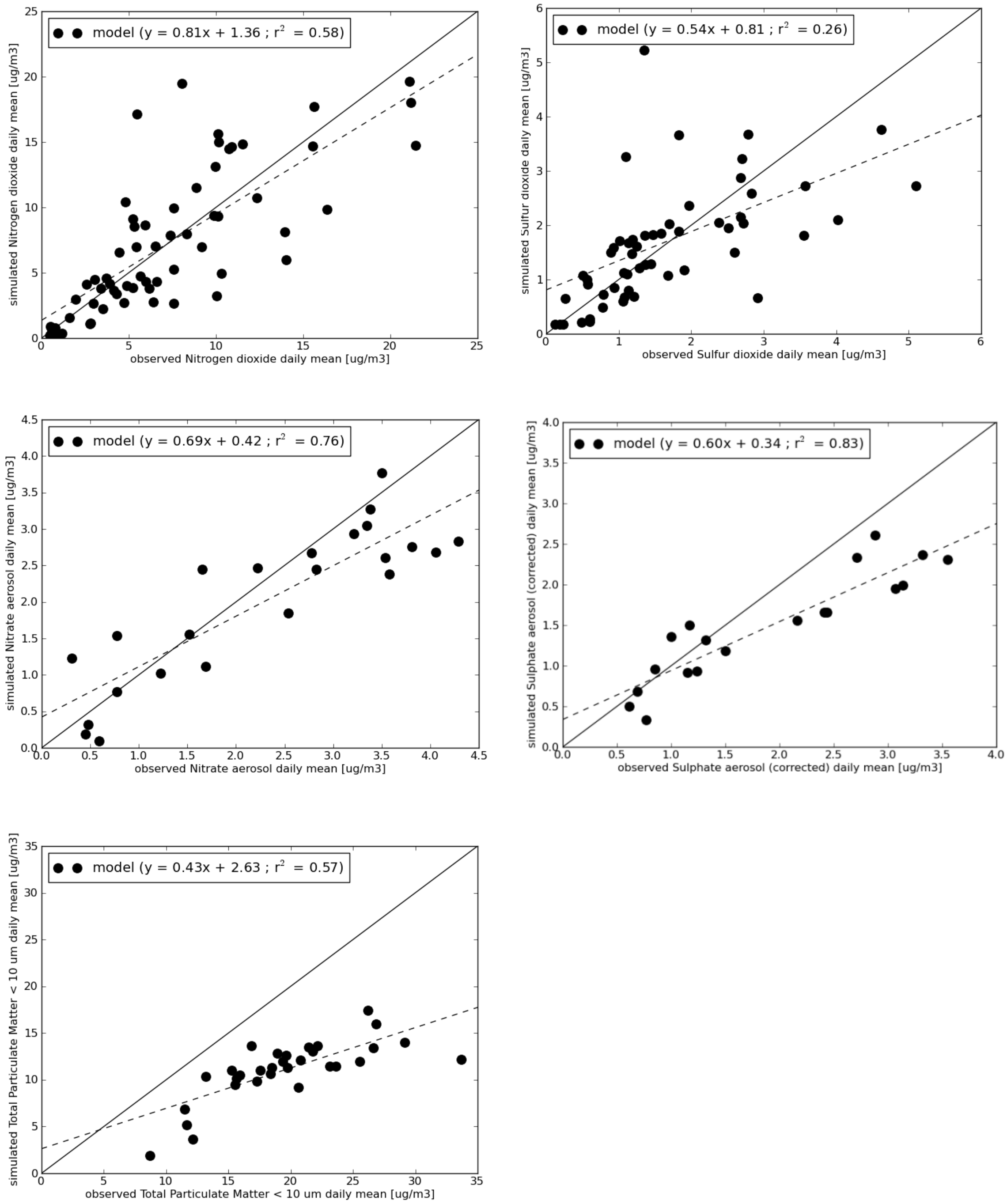

Figure 1 Comparison of modeled NO_2 (top left), SO_2(top right), NO_3 (middle left), SO_4 (middle right) and PM_{10} (bottom) with observations from the EMEP network (Tørseth et al., 2012)

The performance of version 1.8 of LOTOS-EUROS is validated against measurements from regional background stations of the EMEP network[57] for the year 2006. In Figure 1 the annual mean modelled concentrations of SO_2, SO_4, NO_2, NO_3 and PM_{10} are compared to observations. In general the model shows skill in describing the spatial distributions of these pollutants. For the primary species SO_2 and NO_2 there is no indication for a systematic bias between the model and observations. The model strongly over- or underestimates observed concentrations for a few stations, causing a lower coefficient of determination for NO_2 (r^2 = 0.58) and SO_2 (r^2 = 0.26) in comparison to the secondary component sulfate and nitrate (r^2 = 0.83 and 0.76, respectively). For particulate sulfate and nitrate, observed concentrations at the stations with the highest levels are underestimated by LOTOS-EUROS by about 25% and 33%, respectively. Particulate matter concentrations are systematically underestimated by the model by about 40% on average, with r^2 = 0.57. The reason for the underestimation of total PM10 is that not all PM components, e.g. mineral dust and secondary organic aerosol, are included in the model system. On average for all stations, temporal correlations (R^2) of daily averages for the four substances are between 0.43 – 0.57.

Importance of the power sector for air pollutant emissions and concentrations in the several scenarios.

Table 3: Emissions from the power sector and their share of total emissions for a number of countries for the baseline and the 50/50 scenario (also emission totals for the 'low emission' scenario)

		NO_x		PPM_{10}		SO_2	
Country		reference	50/50	reference	50/50	reference	50/50
CZE	emissions from power sector(kton)	94387	45997	4158	2026	137352	66935
	% of total emissions	34.0	16.5	12.1	5.9	62.8	30.6
DEU	emissions from power sector(kton)	241796	156499	10527	6814	215872	139721
	% of total emissions	15.6	10.1	5.0	3.2	38.4	24.9
FRA	emissions from power sector(kton)	106942	30946	8626	2496	120988	35010
	% of total emissions	9.1	2.6	1.7	0.5	26.1	7.6
NLD	emissions from power sector(kton)	44987	33965	294	222	8454	6383
	% of total emissions	12.9	9.8	0.7	0.6	13.6	10.3
POL	emissions from power sector(kton)	289493	176242	25800	15707	644469	392350
	% of total emissions	40.2	24.5	8.9	5.4	52.7	32.1

Table 3 shows the reduction in emissions from the power sector for the 50/50 solar wind scenario (the same emission totals were used in the 'low emission' scenario) compared to the reference scenario for NO_x, primary PM_{10} and SO_2. This table shows that the share of emissions caused by the power sector differ greatly per substance and country. In general, SO_2 emissions have the highest contribution from power plants, especially in countries with many coal-fired power plants (Czech Republic, Poland). In the 50/50 scenario, emissions from power plants are lower for all countries and substances and take up a smaller share of the total emissions (note that emissions from other sectors were kept constant). The reduction in emissions is strongest for France, where relatively little electricity is produced from fossil fuels as France has many nuclear power plants. Installing a large share of renewables at the cost of fossil fuel power plants therefore causes a larger relative reduction in power plant emissions than for e.g. the Netherlands, where in the current electricity mix fossil fuels are much more dominant. For the maximum wind scenario, the trends in annual emissions are the same as for the 50/50 scenario.

Figure 2 shows the contribution of power plants to the annual average concentration of fine sulfate aerosol for all four simulations. This figure shows that a 40% reduction of power plant emissions causes a reduction in ambient fine sulfate, mostly in Eastern Europe where coal is an important fuel for power plants. A 40% reduction of power plant emissions reduces sulfate concentrations from the power sector in this region reduction by on average around 35%. The effect of using more realistic time profiles for power plant emissions for the 50/50 solar wind case can be seen by comparing the top right and lower left panel of this figure. This shows that part of the concentration reduction achieved by reducing the emissions is canceled out by incorporating the shift in the temporal emission characteristics. When the more realistic time profiles are used, the annual average concentrations are up to 20% higher than using the default time profiles for power plants. Using the time profiles calculated for the scenario with maximum deployment of wind energy, the effect of using realistic time profiles is even larger: half of the reduction in concentration because of the lower emissions from the power sector is canceled when the time profiles are adapted.

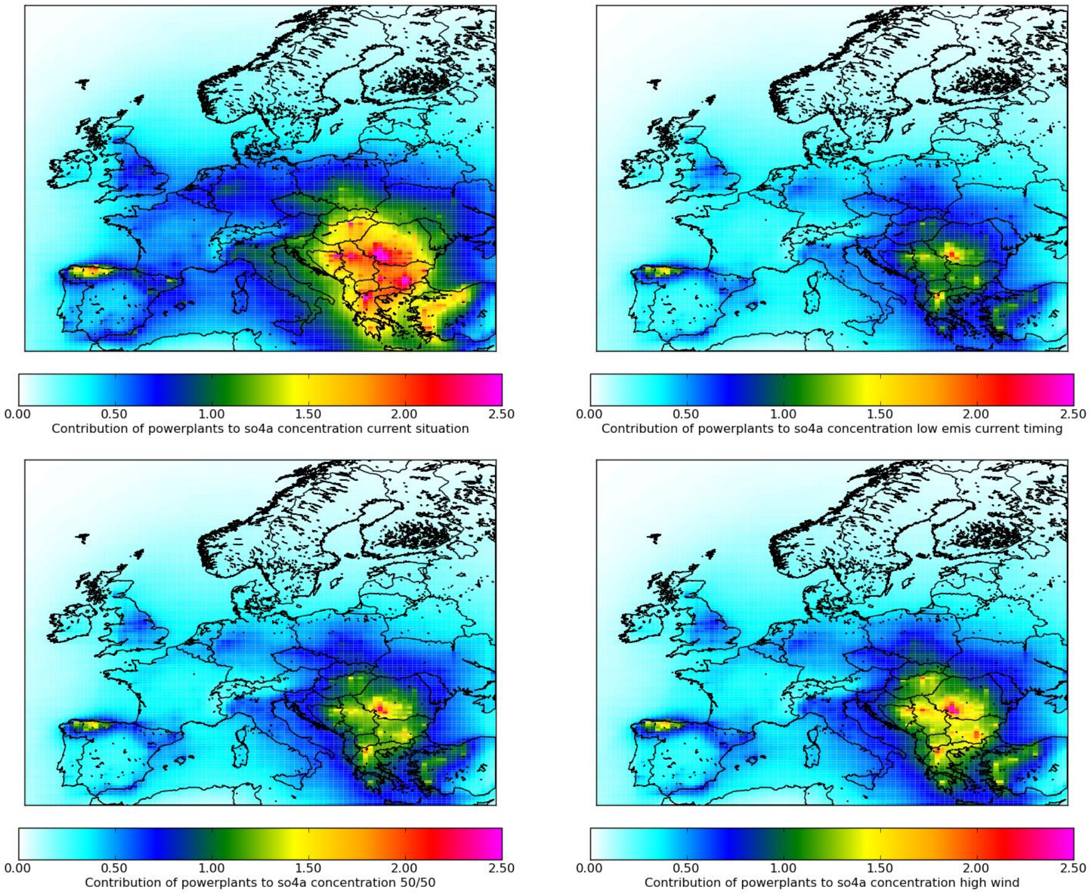

Figure 2 Annual average concentration of sulfate particulate matter (in µg/m^3) for the current situation (top left), low emissions scenario (top right), 25% renewables with equal solar and wind (bottom left) and 25% renewables with maximum wind (bottom right).

Figure 3 shows that these trends are also observed for fine nitrate aerosol, displaying the contribution of power plants to the annual average concentration of this substance. Comparing Figures 2 and 3 show that power plants have an impact on fine nitrate concentrations across the continent, where fine sulfate concentrations due to (coal fired) power plants are centered in Eastern Europe and North-West Spain. This can be explained by the lower dependency of NO_x emissions on fuel mix used in a country.

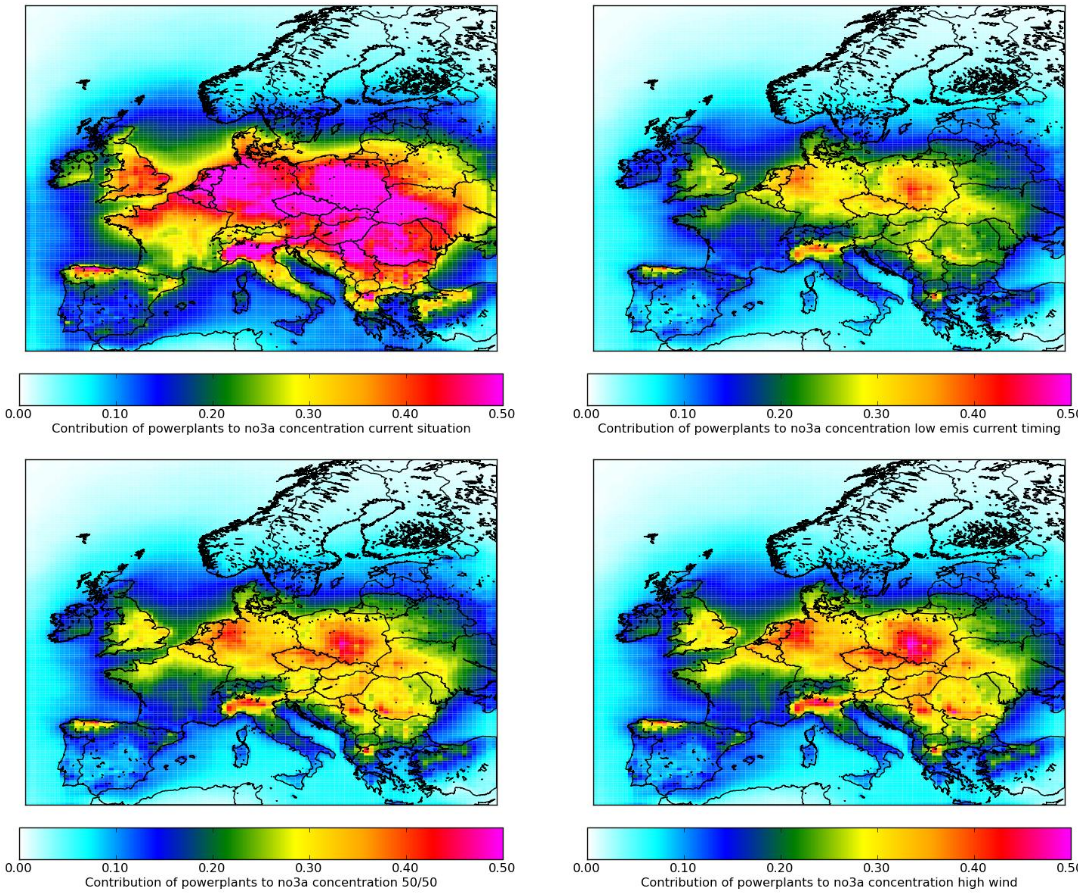

Figure 3 Annual average concentration of nitrate particulate matter (in µg/m^3) for the current situation (top left), low emissions scenario (top right), 25% renewables with equal solar and wind (bottom left) and 25% renewables with maximum wind (bottom right).

Source receptor relations

Next, the effect of timing of power plant emissions on source receptor relations is investigated. The source receptor relations used in this study are country-to-country relations. The impact of selected countries (Table 1) was averaged for all the countries in the exercise. E.g., the concentrations due to the Netherlands in Germany were calculated as the mean over all cells that cover Germany. Cells containing borders were weighted according to the surface area of the countries in the cell.

Figure 4 shows the effect of SO_2 and NO_2 emissions from German and Czech power plants on the concentrations of SO_2, sulphate aerosol and NO_2 for ten European countries. The figure shows the average concentration due to the German/Czech power sector to ten receptor countries, divided by the total emissions (SO_2 for SO_2/SO_4 concentrations; NO_x for NO_2 concentrations) from Germany/Czech republic in each scenario. The result is a measure for the 'effectivity' of emissions in terms of resulting air pollution. As Figure 4 illustrates, reducing SO_2 emissions without changing the time profile yields slightly higher concentrations of SO_4 per unit of SO_2 emission and slightly lower SO_2 concentrations per unit of SO_2 emission in most receptor countries. This is an effect of the inherent non-linearity of the chemistry processes in the atmosphere. Looking at the effect of the change in time profile (compare second and third bars for each country) shows that this increases the effectivity of SO_2 emissions from German/Czech power plants for all receptor countries. This effect is strongest for countries close to the source country and can amount to 40% of the original pollutant/emission ratio (e.g. for sulphate aerosol from Czech power plants). For the high wind scenario, the concentration per unit emission also increases compared to the baseline scenario. Note that the impact for certain receptor countries is larger than others, for example the change in impact of the German power sector is larger for the Netherlands than for Poland.

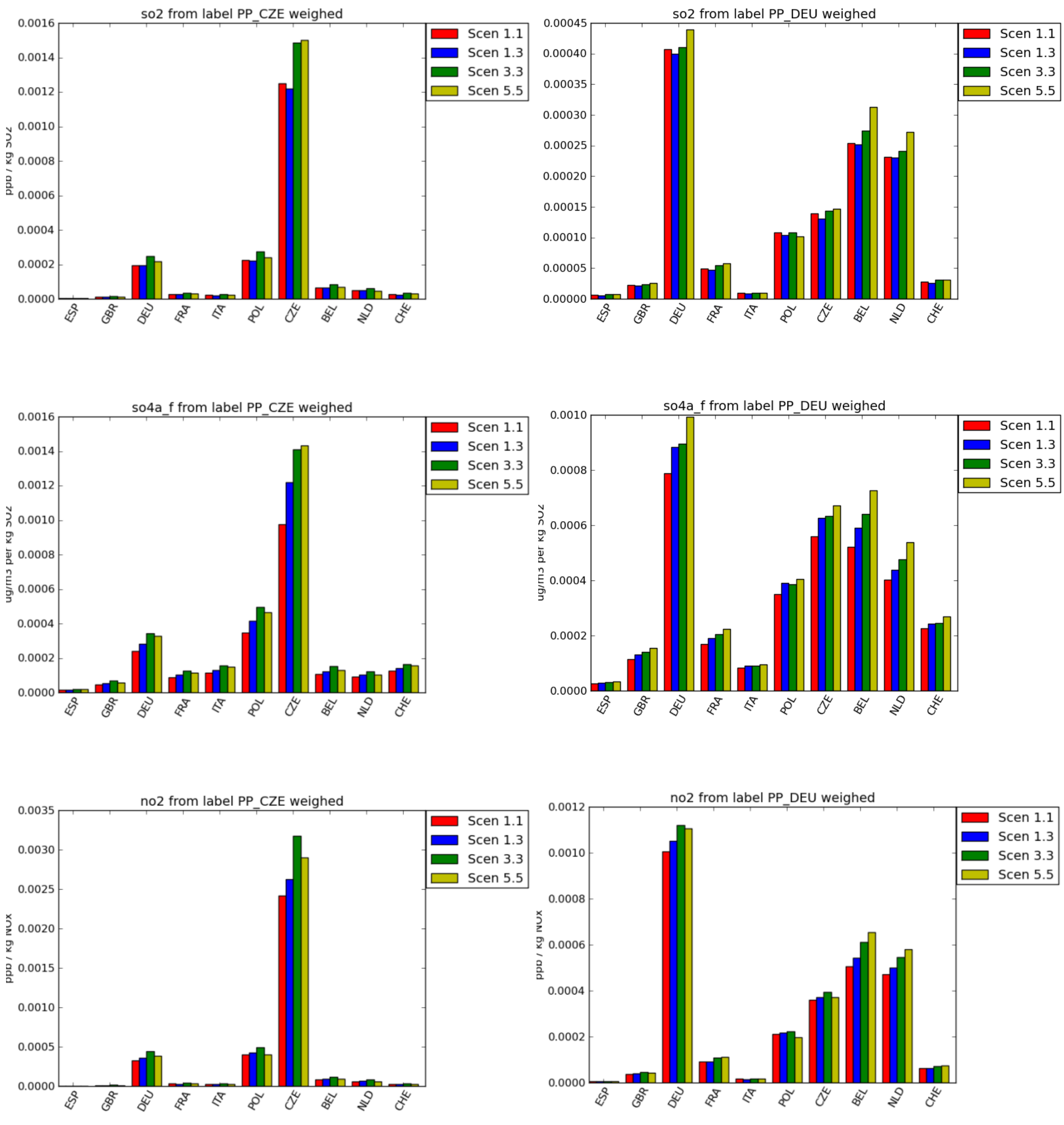

Figure 4 Effect of changing emission quantity and time patterns on SO_2 (top), SO_4 (center) and NO_2 (bottom) concentrations attributed to the Czech (left) and German (right) power sector. (in ppb /kg of emission for SO_2 and NO_2 and $ugSO_4/m^3/kg$ of SO_2 emission for SO_4). Scen1.1 = baseline scenario; Scen1.3 = 'low emission' scenario with emissions of the 50/50 scenario and time profiles of the baseline scenario; Scen3.3 = 50/50 solar/wind electricity, together making up 25% renewable electricity; Scen5.5 = 25% renewables using maximum wind electricity.

For SO_2 emissions, the power sector can be quite dominant, especially in Eastern Europe as SO_2 is mainly emitted during coal combustion. For NO_x, other sectors like transport are also important emitters. The effect of the non-linear chemistry due to the emission reduction for NO_2 'effectivity' is up to 8%, while the effect of the change in emission timing causes up to 23% higher NO_2 concentrations per unit of NO_x emission from power plants. Overall, the increase in effectivity of power plant emissions for the high renewable scenarios is found for all substances and also for other countries, illustrating that the effect found here is systematic.

For a selection of countries, the ratio of concentrations across the domain for the 50/50 and the 'low emission' scenarios weighed with the respective emissions of the substance or its precursor are shown in Figure 5. The effect of the change in timing is larger for the secondary substances (NO_3 and SO_4) than for the primary substances. Secondary inorganic aerosols (SIA) in general have a longer lifetime than its precursors (SO_2 and NO_2 shown here) which are quickly removed via chemical reactions and might not be present long enough to accumulate in the atmosphere. The concentration of SIA components could therefore be more sensitive to weather conditions and e.g. mixing layer height. The impact of emission timing on average concentrations for France and the Czech republic are more pronounced than for the other countries. The remaining emissions from fossil fuel combustion in the power sector for the renewable energy scenarios are very small for France because it has a large share of nuclear power (compare to Table 1). Therefore the fluctuations in emissions from the power sector is larger than for countries for which fossil fuel power stations are also still needed for the base load. For the Czech republic the same reasons apply, although the effect is less pronounced because nuclear energy is less important than in France. Although there are differences across species and countries, this figure illustrates while the magnitude of the effect depends on source country and substance, it is found everywhere.

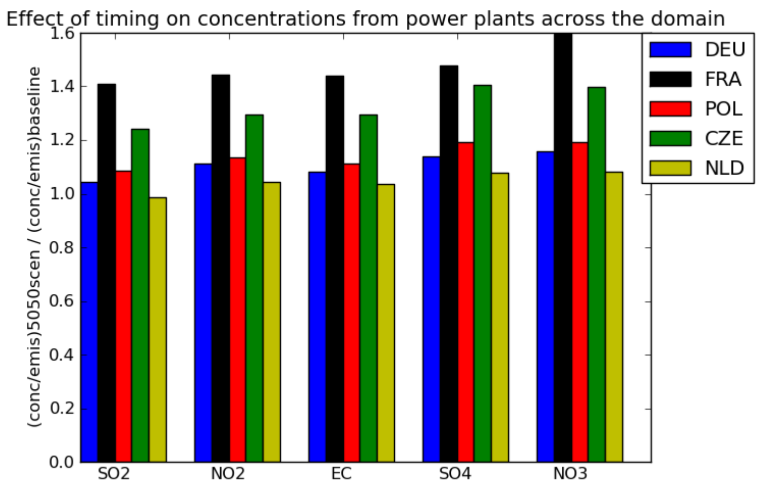

Figure 5 Ratio of concentrations of air pollutants in the 50/50 and 'low emission' scenario from the power sector weighed with the emissions in Germany, France, Poland, Czech Republic and the Netherlands

The top panels of Figure 6 display the contribution of the power sector to sulfate concentrations in winter (left) and summer (right) for the 'low emission' scenario. The sulfate concentrations due to fossil fuel combustion in power plants is higher in summer in the regions in which coal-fired power plants are commonly used (a factor 2 in South-Eastern Europe and a factor 6 in Northern Spain) whereas for the rest of Europe the sulfate levels due to the power sector are about the same for summer and winter. The effect of emission timing is shown in the bottom panels of Figure 6, where the difference between the 50/50 scenario and the 'low emission' scenario is shown. In the summer, concentrations due to power consumption are 10% higher in South-Eastern Europe because of the adjusted time profiles. In winter, this increase in concentration is 20% in both South-Eastern Europe and the Atlantic coastal region The absolute difference between the two runs is slightly larger for the summer in South-Eastern Europe, but for the Atlantic coast the difference is larger in winter. Looking at the monthly average time profiles for the power sector (Figure 7, top panel) explains why the increase in concentrations is larger in winter than in summer: the seasonal variation in the default profile is much flatter than the time profiles in the 50/50 scenario, which shows a larger emission intensity in winter. Actually, when considering this figure the difference between the 'low emission' and the 50/50 scenario in summer would be expected to be negligible, which it is not the case. The increase in concentration can be explained by the distribution of emissions over the day (Figure 7, bottom panel). In the 'low emission' scenario, the emissions peak when the demand is highest, i.e. around noon. For the 50/50 scenario, the emission timing is adjusted to take into account the hourly production of renewable electricity and the emission peak from fossil fuel based power generation shifts to the night hours. During the day enough wind and especially solar energy is available to cover (the major part of) the demand. During the night there is of course no solar electricity (remember we did not include electricity storage) and wind speeds are generally lower than during the day, so fossil fuel-based electricity is needed to meet the demand. As during the night the atmosphere is generally more stable (because of lower mixing layer height, lower wind speeds, sometimes inversion), the average concentration increases when a larger part of emissions is taking place during the night.

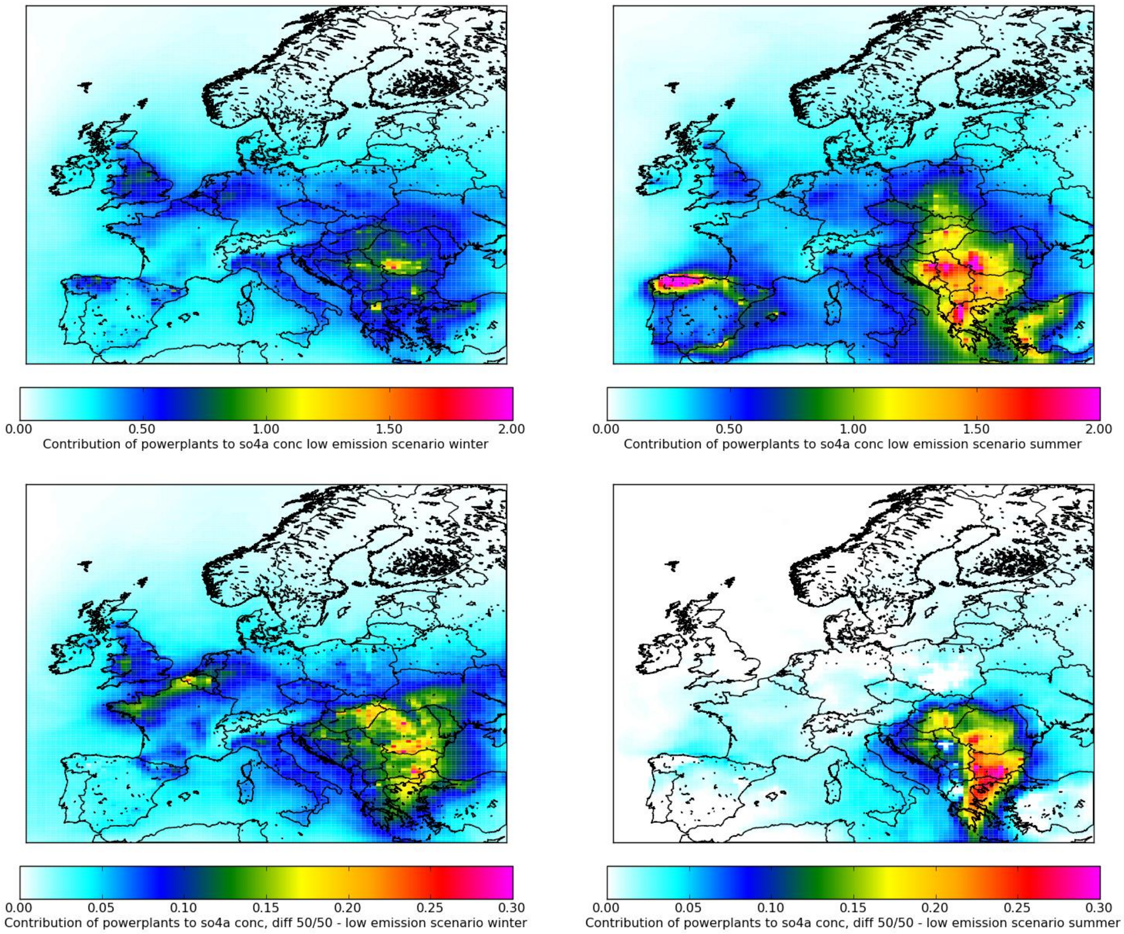

Figure 6 Seasonal average concentration (in µg/m^3) of sulfate aerosol from power plants in the low emission scenario (top) and the difference of the 50/50 scenario and the low emission scenario (bottom). Left: winter (December, January, February); Right: summer (June, July, August).

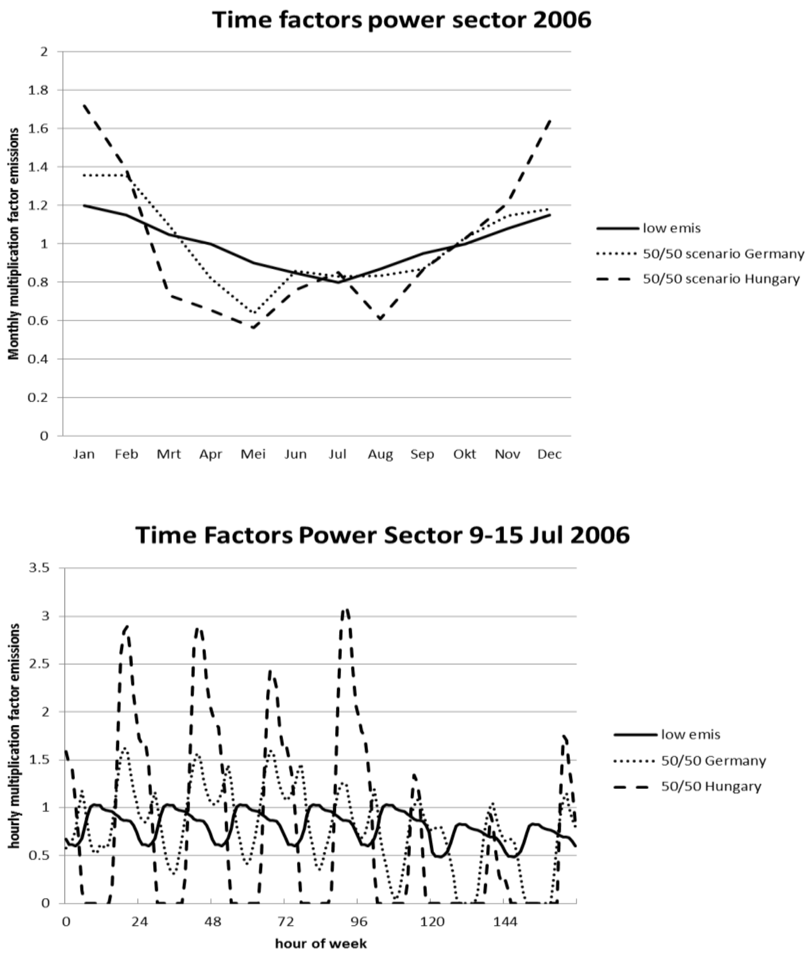

Figure 7 Multiplication factors for power plant emissions for each month (top) and for each hour in a summer week (bottom) in Germany and Hungary. In the low emission scenario, Germany and Hungary have identical time profiles.

5. Discussion and conclusions

This study explored for the first time the consequences for air quality of a shift in the temporal variability of fossil fuel combustion in the power sector induced by an increasing use of renewable energy resources. To isolate the impact of emission timing, two scenarios including the impact of solar and wind energy were developed and compared to a base case in which the timing was kept constant. The results showed that for all species considered the concentration per unit of emission from the power sector is larger when fossil fuel-based power plants operate mainly as backup capacity in an energy system with a significant share of renewable electricity. The impact was found to be larger for secondary species than for primary components with increases of effectiveness of up to 40% and 20%, respectively. Hence, the shift in temporal variability may cause a smaller improvement in air quality than calculated with the current practice which assumes constant time profiles. The reasons for the observed behavior is a larger seasonal variation in emission strength with maxima under winter time stagnant conditions. In addition, in summer emissions from the power sector shift from a day time maximum to a night maximum causing less dilution. The results of this study may have some important consequences and need to be verified with more detailed studies as discussed below.

It has been posed by several authors that the emission data used in CTMs are too static[58,59]. However, the impact of emission time profiles on modeled pollutant concentrations and model performance and results has been given little attention in the past. De Meij et al.[19] found that in the global TM5 model the diurnal and day of the week profiles are only important for NOx, NH3 and aerosol nitrate, whereas for all aerosol species (SO4, NH4, POM, BC) the seasonal emission variability was important. In line with these results, improved temporal variability for road transport has shown to improve model performance of NO2 concerning diurnal and week cycles[17,60,61]. Hence, there are some strong indications that improving emission variability may improve model skill. The calculation of anthropogenic emissions in CTMs follows the same procedure since the early nineties. Annual emission totals are spatially distributed using proxy maps and point source information. These spatially distributed inventories are combined with static time profiles per sector to calculate the emission of air pollutant at each hour of the simulation. Skjøth et al.[62] moved away from this practice for the agricultural sector and found an improvement in CTM performance by applying a dynamic ammonia emission model which accounts for local agriculture management and local climate. Mues et al.[17] showed that temperature dependent emissions for domestic heating improves model performance. Based on our results we recommend to also build a detailed emission model of energy sector to be able to assess impacts of an energy transition in detail, especially considering the anticipated electrification of the transport and industry sectors which will cause emissions from the power sector to be larger in both relative and absolute terms.

When assessing the impact of a shift in emission timing on air pollution levels, it is important to know how well a CTM explains variability in concentrations over time and space in the current situation. Many CTMs, including LOTOS-EUROS, underestimate the variability of air pollutant levels in general and specifically as a function of meteorology[52,63]. The underestimation of variability in concentrations is mainly caused by the underestimation of concentration peaks[52,59]. These peak episodes mainly occur during stagnant meteorological conditions, during which most fossil fuel power plant emissions remain in our scenarios. Therefore, assuming that the too simplistic representation of the temporal variability of emissions is not the main reason for the underestimation of the peak concentrations the increase of concentration per unit of emission from power plants because of the change in emissions timing might well be underestimated. Therefore, a reanalysis effort for the last 1-2 decades is necessary to determine the impact of an temporally explicit emission model (containing all sectors) to assess the sensitivity of the model results to the emission description.

Considerable shifts in the diurnal cycle of NOx emissions may also impact ozone formation. Previous studies found a significant increase in model performance when considering emission profiles for the day-of–week and in the diurnal cycle compared to a simulation with constant emissions[64]. Inclusion of a day of the week emission profile led to successfully capturing the higher observed ozone concentrations in the weekend than during weekdays by the CMAQ model[60]. The ozone formation potential per unit emission is dependent on the ratio between anthropogenic and biogenic VOCs and NOx as well as meteorological conditions[65]. Hence, the ozone formation potential per unit emission is likely to change considerably when emissions shift from day to night time due to the different fate of NOx during day and night time chemistry[66]. Unfortunately, our source apportionment module is not suited for tracing ozone origin, so we could not separate the impact of the power sector from the other important NOx and VOC emitting sectors. Hence, for the assessment of the impact of the power sector on future ozone levels a dedicated scenario study remains to be performed.

The scenarios developed in this study were not meant to be a realistic representation of a possible future, but only as an instrument to explore the impact of a shift in time profiles of emissions from power plants in Europe on air quality. Three important assumptions were made that impact the results of this study. First, no storage and trade of electricity was accounted for. Also, hydroelectricity production is assumed to be constant over the year, whereas in reality the electricity production from this source can be regulated almost instantly and water reservoirs can even be used to store excess electricity. Including these factors would partly counterbalance the intermittent character of wind and solar energy and balance the timing of fossil fuel combustion emissions through the year and throughout Europe. Secondly, the electricity demand was assumed not to change in quantity and time pattern. The electricity demand in Europe is anticipated to increase over the coming years, meaning that with the assumed amount of electricity production from renewable sources more fossil fuel-based electricity will be needed than estimated here. When electrification of e.g. the transport sector is considered, the time pattern of electricity demand might change as well. This will not only impact the time and quantity of electricity production but will also increase the relative importance of the power sector in terms of emissions compared to other sectors. The third important assumption is that the fuel mix of power plants is not changed. In reality the response to a decrease in fossil fuel electricity demand will be the shutdown of older power plants. Also, gas power plants can in general be switched on and off more quickly than coal fired plants. Therefore, the fuel mix is anticipated to change vary with electricity demand and meteorology. The latter is expected to more relevant for emissions of sulfur dioxide than nitrogen oxides. Future scenario studies should test the importance of these major assumptions.

Within integrated assessment models such as GAINS[20] the SRRs are at the core of the development of cost effective mitigation strategies for climate change and air pollution. They are assumed to be linear in the optimization simulations. Currently, SSRs are calculated by reducing one by one the pollutant emission total by 15% for each country in Europe assuming no change in emission timing[22]. The assumption that the SRRs behave linearly is assumed to hold when the emission change is less than 15% of the total annual emission[21], as for larger changes non-linear effects in atmospheric chemistry cannot be neglected anymore. In our simulations, the impact of a shift in temporal profiles is larger than the non-linear effect induced by a change in the chemical regime (by a 40% reduction in power sector emissions). This may mean that for system changes that involve shifts in the temporal and geographical profile of emissions, SRRs may be non-linear for much smaller changes in the total emissions than currently assumed. Hence, in case our results are validated in more extensive studies, refined SRRs for assessing the impacts of an energy transition appear to be needed.

Recent research suggests that the co-benefits of climate change policy for air quality are much larger than vice versa[67]. An important consequence of our results is the implication that the estimated co-benefits from climate change policies for air quality might be too optimistic when impacts on emission timing are not considered. It should directly be noted that this is probably not important in case the projected power sector emissions are marginal compared to the current situation (e.g. a very large share of renewable electricity or very effective emission control). Our simulations are more representative for the transition phase towards a renewable power sector. As a fully renewable energy system is at least a few decades away, the outcomes of this study may be relevant for anticipating and monitoring the impact of policies for the next 20-40 years. Note that we have addressed the impact on ground level air quality concentrations only and our results are therefore cannot directly translated to an impact on radiative forcing. Air pollutants as particulate matter and ozone are considered to be important short lived climate forcers. We speculate that the impact we illustrated for secondary sulfate may also be relevant of future climate impacts of regional aerosol distributions, in Europe or elsewhere in the world. In short, to improve our capability to forecast the levels and impacts of air pollutants during a transition to a renewable energy system, the representation of fossil fuel combustion in CTMs needs to be more detailed.

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