Response of fine particulate matter to changes of emissions in Europe

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1. Abstract

PMCAMx-2008, a three dimensional chemical transport model (CTM), was applied over Europe in order to quantify the changes in fine aerosol ($PM_{2.5}$) mass concentration in response to different emission reductions. The effects of 50% reduction emissions of sulfur dioxide (SO_2), ammonia (NH_3), oxides of nitrogen (NO_x), anthropogenic volatile organic compounds (VOCs), and anthropogenic primary OA (POA) was investigated. Two simulation periods were used, summer 2008 and winter 2009 to examine the effectiveness of the control strategies. Summer results were based on a hot period during May 2008 while winter simulation was based on results during a late winter and early spring period (February/March 2009). The reduction of NH_3 emissions reduces $PM_{2.5}$ up to 5 μg m^{-3} and 1.5 μg m^{-3} during summer and winter respectively, mainly because of reductions of ammonium nitrate. Reduction of SO_2 leads also to a decrease of $PM_{2.5}$ in both periods, having a significant effect over the Balkans region (up to 1.6 μg m^{-3}) during summer. The reduction of NO_x emissions results in a decrease of $PM_{2.5}$ concentrations (up to 3.4 μg m^{-3}) during summer but it is insufficient for reducing $PM_{2.5}$ levels during winter. The reduction of VOCs and anthropogenic primary OA emissions reduce total OA levels by less than 1 μg m^{-3} in both periods, having a negligible effect on inorganic aerosol components.

2. Introduction

Atmospheric particles have adverse effects on human health and have been implicated in the formation of acid rain and acid fogs, visibility reduction and changes of the energy balance of the planet. Ozone, particulate matter less than 2.5 μm in size ($PM_{2.5}$) and other pollutants either organic or inorganic are subjected to a complex series of common emissions, physical and chemical transformations. Consequently improving air quality requires understanding of how the emissions reductions of one pollutant can lead to changes (either positive or negative) in the concentration of other pollutants. A three-dimensional chemical transport model (PMCAMx-2008) can estimate these source-receptor relations because it directly links emissions to $PM_{2.5}$ concentrations through detailed descriptions of the physics and chemistry of the atmosphere.

3. PMCAMx-2008 Description

PMCAMx-2008 (Murphy and Pandis, 2009; Tsimpidi et al., 2010; Karydis et al., 2010) uses the framework of the CAMx air quality model (Environ, 2003) which simulates the processes of horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, and gas-phase chemistry. Three detailed aerosol modules are used: inorganic aerosol growth (Gaydos et al.,2003; Koo et al., 2003), aqueous phase chemistry (Fahey and Pandis, 2001) and SOA formation and growth (Koo et al., 2003). In addition, the model includes a state-of-the-art organic aerosol module which is based on the volatility basis set framework (Donahue et al., 2006; Stanier et al., 2008) treating both primary and secondary organic components to be semivolatile and photochemically reactive.

4. Model Application

PMCAMx-2008 is applied over Europe covering a 5400 × 5832 km^2 region with 36 × 36 km grid resolution and 14 vertical layers covering approximately 6 km. The necessary inputs to the model include horizontal wind components, vertical diffusivity, temperature, pressure, water vapor, clouds and rainfall, all created using the meteorological model WRF (Weather Research and Forecasting). In addition anthropogenic (Visschedijk et al., 2007) and biogenic hourly emission gridded fields were developed for the European domain for gases and primary particulate matter (Table 1).

Table 1. Emission mass totals for gaseous and PM_{10} species (in ktons/month) for the European domain during summer 2008 and winter 2009

Species		CO	NO_x	SO_2	NH_3	NMVOC	NO_3^-	Sulfate	NH_4^+	EC	OC	Na^+	CL^-
Summer	Anthropogenic	3682	1461	1675	501	1291	-	-	-	59	82	-	-
	Biogenic	1207	46	3	11	2890	7	1309	1	16	93	496	89
Winter	Anthropogenic	5002	1531	1961	474	1334	-	-	-	77	126	-	-
	Biogenic	80	5	59	-	381	-	2933	-	-	12	11219	2016

5. Model Evaluation

The averaged ground-level concentration predictions during the summer period were evaluated against high time resolution aerosol mass spectrometer (AMS) measurements taken from various sites in Europe during the EUCAARI intensive periods (Fountoukis et al., submitted). The comparison of the model predictions with the measurements was encouraging (Fig 1). The model reproduced more than 87% and 70% of the hourly averaged data within a factor of 2, for PM_1 OA and sulfate respectively (Table 2).

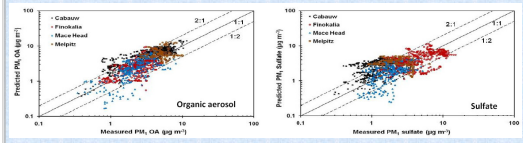

Figure 1. Comparison of predicted vs. observed PM_1 organic matter and sulfate concentrations (μg m^{-3}) from 4 measurement stations during the EUCAARI summer 2008 campaign. Each point corresponds to a 1-hour average value. Observed data represent AMS measurements.

Table 2. Prediction skill metrics of PMCAMx-2008 against AMS hourly ground measurements from 4 stations (Cabauw, Finokalia, Mace Head, Melpitz) during the EUCAARI summer 2008 campaign.

PM_1	Mean Observed (μg m^{-3})	Mean Predicted (μg m^{-3})	MB (μg m^{-3})	MAGE (μg m^{-3})	NMB (%)	NME (%)	FBIAS	FERROR	Percent within a factor of 2
OA	3.3	3.0	-0.4	1.0	-11	30	-0.1	0.3	87
Sulfate	2.8	2.9	0.1	1.3	3	47	0.1	0.4	70
Nitrate	1.7	2.8	1.1	1.8	67	105	0.4	0.8	28
Ammonium	1.5	1.7	0.3	0.8	23	58	0.2	0.5	67

6. Reduction of NH_3 emissions by 50%

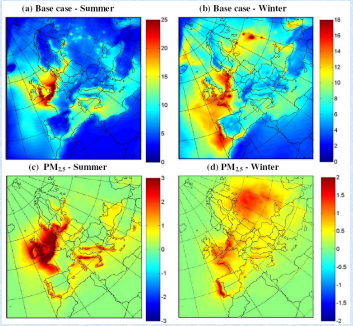

Figure 2. Predicted concentrations and concentration change in average ground-level $PM_{2.5}$ concentrations (μg m^{-3}) for (a,b) base case and (c,d) after a 50% reduction of NH_3 emissions during summer 2008 and winter 2009. A positive value corresponds to a decrease.

$PM_{2.5}$ is predicted to decrease in both periods mainly due to reductions of ammonium nitrate.

In summer, the reduction of $PM_{2.5}$ is highest over a wide area from United Kingdom to Denmark with a predicted maximum decrease of 5.1 μg m^{-3} (22.5%) in south England (Fig 2c). A significant decrease of $PM_{2.5}$ is also predicted in Albania (1.8 μg m^{-3} or 15%) and in western Italy (2 μg m^{-3} or 18%). The average reduction of $PM_{2.5}$ is around 0.5 μg m^{-3} (5.5%)

During winter, $PM_{2.5}$ is predicted to decrease up to 1.5 μg m^{-3} (10%) in western Portugal (Fig 2d). A significant decrease is also predicted in Russia (1.3 μg m^{-3} or 12%). On average $PM_{2.5}$ decreases around 0.4 μg m^{-3} (4%).

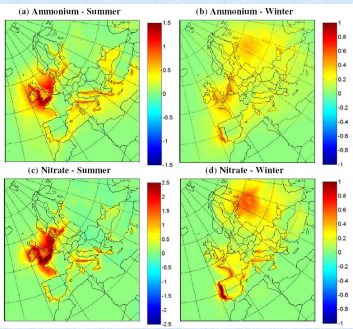

Figure 3. Predicted change (μg m^{-3}) in ground-level concentrations of (a,b) ammonium, and (c,d) NO_3^- after a 50% reduction of NH_3 emissions during summer 2008 and winter 2009. A positive value corresponds to a decrease.

In summer the ammonium reduction exceeds 10% everywhere. The highest reduction is predicted over the area from United Kingdom to Denmark with a predicted maximum decrease over United Kingdom (approximately 1.3 μg m^{-3} or 35.5%) (Fig 3a). Nitrate has a similar trend with the highest reduction predicted in South Wales around 3.1 μg m^{-3} (50%) (Fig 3c).

In winter, nitrate is predicted to decrease up to 1.1 μg m^{-3} (52%) in western Portugal (Fig 3d). The respective highest reduction of ammonium is approximately 0.7 μg m^{-3} (39%) (Fig 3b).

7. Reduction of NO_x emissions by 50%

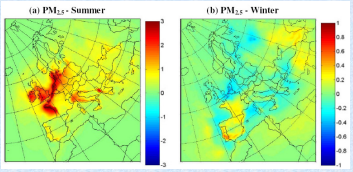

Figure 4. Predicted change (μg m^{-3}) in ground-level concentrations of $PM_{2.5}$ after a 50% reduction of NO_x emissions during (a) summer 2008 and (b) winter 2009. A positive value corresponds to a decrease.

During summer, $PM_{2.5}$ is predicted to decrease in the entire model domain. The reduction of $PM_{2.5}$ is highest over a wide area from United Kingdom to Denmark with a predicted maximum decrease 3.4 μg m^{-3} (17%) in the northern coast of France (Fig 4a).

In winter, the response of $PM_{2.5}$ is different. $PM_{2.5}$ either increases (due to increase of organics and sulfate) or remains constant except a slight decrease mainly in Spain, Portugal and France. (up to 0.4 μg m^{-3}) (Fig 4b)

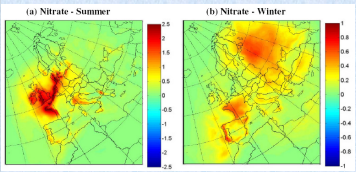

Figure 5. Predicted change (μg m^{-3}) in ground-level concentrations of NO_3^- after a 50% reduction of NO_x emissions during (a) summer 2008 and (b) winter 2009. A positive value corresponds to a decrease.

Nitrate is predicted to decrease in both periods. During summer nitrate decreases around 0.3 μg m^{-3} (42%) on average with a predicted maximum decrease 2.9 μg m^{-3} (47%) in the northern coast of France (Fig 5a).

In winter, the highest reduction of nitrate is predicted in northern Spain (approximately 0.8 μg m^{-3} or 30%) (Fig 5b). On average nitrate decreases around 0.2 μg m^{-3} (34.5%).

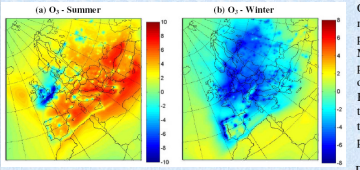

Figure 6. Predicted change (ppb) in ground-level concentrations of O_3 after a 50% reduction of NO_x emissions during (a) summer 2008 and (b) winter 2009. A positive value corresponds to a decrease.

O_3 is predicted to decrease in most of the model domain during summer with a predicted maximum decrease over the Mediterranean region close to Cyprus (8 ppb or 18%) (Fig 6a). However an increase of O_3 is predicted over an area from United Kingdom to Denmark (8 ppb or 40%). In this area sulfate and organics are predicted to increase. Increases of a few ppb are also predicted in most urban areas.

During winter O_3 is predicted to increase in most of the model domain. The highest increase, approximately 9 ppb (80%) is predicted in the northeastern coast of Spain (Fig 6b).

8. Reduction of SO_2 emissions by 50%

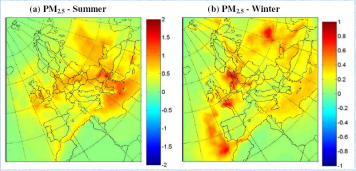

Figure 7. Predicted change (μg m^{-3}) in ground-level concentrations of $PM_{2.5}$ after a 50% reduction of SO_2 emissions during (a) summer 2008 and (b) winter 2009.A positive value corresponds to a decrease.

$PM_{2.5}$ is predicted to decrease in both periods mainly due to reductions of sulfate. In summer, $PM_{2.5}$ decreases significantly over the Balkans region with a predicted maximum decrease 1.6 μg m^{-3} (17%) in south Bulgaria (Fig 7a). The respective decrease of sulfate is 1.3 μg m^{-3} or 40% (23% on average).

In winter, the highest reduction of $PM_{2.5}$ is predicted in northern France (0.7 μg m^{-3} or 5%) and in Denmark (0.9 μg m^{-3} or 7%) (Fig 7b). Sulfate decreases 0.3 μg m^{-3} (15%) on average.

9. Reduction of VOC emissions by 50%

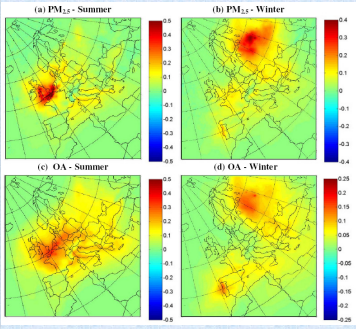

Figure 8. Predicted change (μg m^{-3}) in ground-level concentrations of (a,b) $PM_{2.5}$ and (c,d) total OA after a 50% reduction of anthropogenic VOCs emissions during summer 2008 and winter 2009. A positive value corresponds to a decrease.

In summer, the highest reduction of $PM_{2.5}$ is predicted in northern France (0.5 μg m^{-3} or 2%) (Fig 8a). The respective highest reduction of total OA is approximately 0.3 μg m^{-3} (8%) (Fig 8c).

During winter $PM_{2.5}$ is predicted to decrease up to 0.4 μg m^{-3} (1%) in northwestern Russia (Fig 8b), while the highest reduction of total OA is predicted around 0.2 μg m^{-3} (2%) (Fig 8d).

On average total OA decreases 5% in summer and 3% in winter respectively.

10. Reduction of POA emissions by 50%

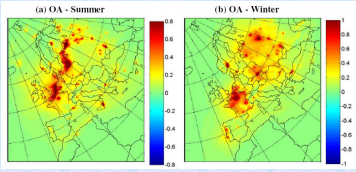

Figure 9. Predicted change (μg m^{-3}) in ground-level concentrations of total organic aerosol after a 50% reduction of anthropogenic POA emissions during (a) summer 2008 and (b) winter 2009. A positive value corresponds to a decrease.

During summer, total OA is predicted to decrease up to 3.6 μg m^{-3} (34%) in southern Finland (Fig 9a), while the highest reduction of total OA in winter is predicted in northern France, close to Paris (approximately 6.7 μg m^{-3} or 48%) (Fig 9b).

On average total OA decreases 6% in summer and 11% in winter respectively.

$PM_{2.5}$ has a similar trend. The predicted decrease of $PM_{2.5}$ is mainly due to reductions of organics

Table 3. Response of the major $PM_{2.5}$ to a 50% reduction of NH_3, NO_x, SO_2, VOCs and POA emissions, during summer 2008 and winter 2009.

Control Strategy		$PM_{2.5}$	Ammonium	Sulfate	Nitrate	Total OA
-50% NH_3	Summer	↓	↓	↓	↓	–
	Winter	↓	↓	↓	↓	–
-50% NO_x	Summer	↓	↓	↓↑	↓	↓↑
	Winter	↓↑	↓	↑	↓	↑
-50% SO_2	Summer	↓	↓	↓	↑	–
	Winter	↓	↓	↓	↑	–
-50% VOCs	Summer	↓	–	–	–	↓
	Winter	↓	–	–	–	↓
-50% POA	Summer	↓	–	–	–	↓
	Winter	↓	–	–	–	↓

Conclusions

1. Reduction of NH_3 is the most effective control strategy for reducing $PM_{2.5}$ in both periods. The NH_3 strategy results in a significant decrease of ammonium nitrate.
2. The NO_x control strategy is sufficient for reducing $PM_{2.5}$ during summer, mainly due to reductions of nitrate and ammonium. Sulfate and organics are also decreased except over an area from United Kingdom to Denmark where they increase. Although nitrate and ammonium are reduced in winter the NO_x reduction is insufficient for reducing $PM_{2.5}$ due to an increase of organics and sulfate.
3. During summer the NO_x control strategy increases ozone over an area from United Kingdom to Denmark and in most of urban areas. Thus, the NO_x reduction seems to be problematic.
4. The SO_2 control strategy is effective for reducing $PM_{2.5}$ during summer, especially over the Balkans region, mainly due to reductions of sulfate. However in both periods the SO_2 reduction leads to an increase of nitrate. Ammonium is slightly decreased in both periods.
5. The reduction of anthropogenic VOCs slightly decreases $PM_{2.5}$ levels in both periods due to reductions of organics. The VOCs control strategy has a negligible effect in inorganic aerosol components.
6. The primary OA control strategy slightly reduces $PM_{2.5}$ in both periods. In winter the reduction of total OA is higher.

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Acknowledgements

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