19th International Conference on
Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
3-6 June 2019, Bruges, Belgium

CHANGES OF AIR QUALITY DUE TO THE REVISED GOTHENBURG PROTOCOL: IMPROVEMENT AND CHALLENGE REMAINS

Jianhui Jiang, Sebnem Aksoyoglu, André S. H. Prévôt

Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, 5232 Villigen, Switzerland

Abstract: We modelled the changes in the European air quality between 2010 and 2020 due to the implementation of the revised Gothenburg Protocol by the chemical transport model CAMx (version 6.50). The emissions in 2020 were projected by using the reference year (2005) emissions and country-specific reduction targets (incl. SO_2, NO_x, NH_3, $PM_{2.5}$ and NMVOCs) of the revised Gothenburg Protocol. The same inputs for meteorology, boundary conditions and photolysis rates as in 2010 were used for 2020. The modelled results for 2010 were evaluated by measurements obtained from the European air quality database (AirBase), which indicated good agreements for ozone, SO_2, NO_x and $PM_{2.5}$. The reduced emissions lead to a decrease in the average concentrations of $PM_{2.5}$, SO_2, and NO_x by 12%, 29% and 26%, respectively, between 2010 and 2020. The highest predicted reduction in country level occurs for SO_2 (64%) in Poland, for NO_x in the UK (44%), and for $PM_{2.5}$ (28%) in Switzerland. The annual average ozone mixing ratios in southern Europe show a slight decrease, while an increase by up to ~3 ppb is observed in the Benelux countries and UK, as the reduced NO_x emissions decrease the ozone titration. However, the maximum daily 8-hour mean ozone concentrations generally decrease in summer when highest ozone formation occurs. Over the 2278 tested stations covering rural, urban and suburban area, the maximum daily 8-hour mean ozone concentrations in summer (June–July–August) are ~2 ppb lower in 2020 than in 2010. Among the simulated pollutants, the $PM_{2.5}$ is identified as a crucial target for future. Although the number of countries having annual $PM_{2.5}$ concentration below the World Health Organization (WHO) guidelines (10 µg m^{-3}) increases from 22 in 2010 to 31 in 2020, the eastern Europe could still have high health risk due to $PM_{2.5}$ in 2020.

Key words: *Air quality model, Gothenburg Protocol, ozone, nitrogen oxides, particulate matter*

INTRODUCTION

Air pollution is one of the leading five health risks worldwide. In Europe, despite considerable reductions in emissions of air pollutants under numerous regulations of pollution control, air pollution remains to be a critical issue in many areas. Due to the complex links between emissions and air quality, the emission reductions do not always lead to a decrease in concentrations, especially for the secondary pollutants ozone (O_3) and particulate matter (PM). While the emissions of O_3 precursors nitrogen oxides (NO_x) and non-methane volatile organic compounds (NMVOCs) decreased by 40% in Europe between 2000 and 2016, a similar trend was not observed for O_3 (EEA, 2018). The observed PM_{10} concentrations decreased across

Europe, however, 79% of the stations did not catch significant trends in 2006 – 2011 for $PM_{2.5}$ (Guerreiro et al., 2014), and 81% of the European population was still exposed to annual mean concentration of $PM_{2.5}$ exceeding the World Health Organization (WHO) air quality guidelines (AQG) in 2015 (EEA, 2018). As an international effort to control air pollution, the Gothenburg Protocol was revised in 2012 to include more stringent emission reduction commitment for 2020, in which the reduction target for $PM_{2.5}$ was added. To investigate the effects of reduced emissions on air quality, we modelled the change in pollutant concentrations in Europe between 2010 and 2020 as a result of the implementation of the revised Gothenburg Protocol.

METHOD

The air quality model CAMx version 6.50 (Ramboll, 2018) was used to simulate the European air quality in 2010 and 2020. The model domain covered Europe (17° W - 39.8° E, 32° - 70° N), with a horizontal resolution of 0.4° × 0.25°. There were 14 terrain-following vertical layers reaching up to 8000 m, with the first layer height ~50 m. The Carbon Bond 6 Revision 2 (CB6r2) mechanism was used for the gas-phase chemistry, and the secondary organic aerosol chemistry/partitioning (SOAP2.1) module was used for the aerosol chemistry.

Most of the model inputs (meteorology, anthropogenic emissions, boundary conditions) for the year 2010 were obtained from the EURODELTA-Trends (EDT) database (Colette et al., 2017), while the biogenic emissions were estimated by MEGAN version 2.1. Ozone column densities were obtained from Total Ozone Mapping Spectrometer (TOMS) data by NASA and photolysis rates were calculated using the Tropospheric Ultraviolet and Visible (TUV) Radiation Model version 4.8. For 2020, the same inputs as 2010 were adopted except for the anthropogenic emissions, which were projected based on the reference year (2005) emissions and country specific reduction targets of the revised Gothenburg Protocol (UNECE, 2014), including species of SO_2, NO_x, NH_3, $PM_{2.5}$ and NMVOCs. The modelled results in 2010 were evaluated using the measurements obtained from the European air quality database AirBase version 7 by the European Environment Agency (Mol and Leeuw, 2005).

RESULTS AND DISCUSSION

Model evaluation

The statistical results between the measured and modelled SO_2, O_3, NO_2, NH_3, PM_{10} and $PM_{2.5}$ in 2010 are shown in Table 1. For O_3 and NO_2, only background rural stations were used in the evaluation to reduce the uncertainties arising from grid resolution. We compared the results with the performance criteria (O_3: MFB $\leq \pm 30\%$, MFE $\leq 45\%$; $PM_{2.5}$: MFB $\leq \pm 60\%$, MFE $\leq 75\%$) given by Boylan and Russell (2006) and EPA (2007), and found all the performance criteria were met. Both $PM_{2.5}$ and PM_{10} were underestimated, largely due to the common issue of under-predicted secondary organic aerosol especially in winter. Overall, the model performance on simulating the major air pollutants was comparable with other modelling studies in Europe (Bessagnet et al., 2016; Ciarelli et al., 2016).

Table 1. Model performance evaluation for gaseous and particle species in 2010. MB: mean bias; ME: mean error; RMSE: root-mean-square error; MFB: mean fractional bias; MFE: mean fractional error.

Species	Number of stations	MB[a]	ME[a]	RMSE[a]	MFB (%)	MFE (%)
SO_2	2054	-0.6	1.2	2.5	-15.2	60.8
O_3	606	6.1	7.1	9.0	18.9	21.6
NO_2	548	-1.8	2.6	4.1	-30.2	46.7
NH_3	9	-0.1	0.1	0.2	10.4	64.4
PM_{10}	1415	-14.5	14.8	21.0	-64.8	66.8
$PM_{2.5}$	432	-4.0	5.0	7.5	-30.0	37.3

[a] Units are ppb for gaseous species and $\mu g\ m^{-3}$ for PM.

Changes in air quality

The reduced emissions lead to significant changes in air quality in Europe between 2010 and 2020 (Fig. 1). The average concentrations of SO_2, NO_x and $PM_{2.5}$ in 2020 decrease by 29%, 28% and 17% respectively. The largest decrease among the major pollutants occurs in SO_2 as a direct consequence of stringent SO_2 emission ceilings (average reduction ratio from 2005 level is 59% for EU according to the revised Gothenburg Protocol). The highest country-level reduction of SO_2 reaches 7.8 ppb (61%) in Poland. The

NO_x concentrations decrease mostly in Benelux countries with a net reduction reached ~10 ppb, while the highest country-level decrease is in UK (44%) due to relatively lower base concentrations in 2010. The decrease in $PM_{2.5}$ largely benefits from the decrease of sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+) concentrations due to the reduced SO_2, NO_x and NH_3 emissions. Elemental carbon and organic aerosol concentrations were also predicted to decrease, but the magnitudes were much lower than the inorganic aerosols in most of the area except for Poland and Po Valley. The highest country-level decrease in $PM_{2.5}$ was found to be in Switzerland (28%), and the major contributor was the reduced SO_4^{2-}.

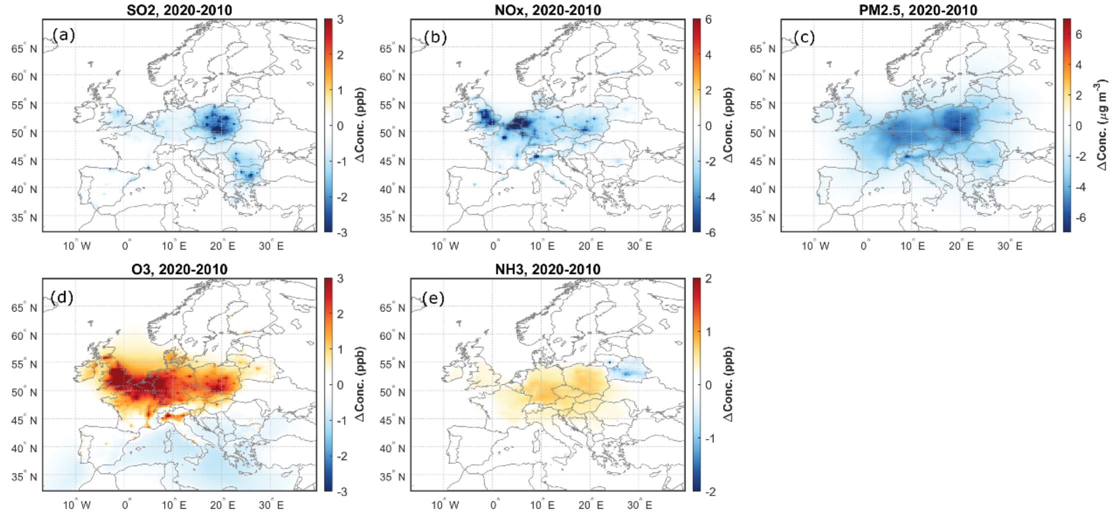

Figure 1. Changes in the yearly average concentrations of the major air pollutants between 2010 and 2020.

The changes in O_3 and NH_3 concentrations were different than the others (Fig. 1d-e). Despite considerable reduction in precursor emissions, the O_3 concentrations increased in central Europe with highest increase up to ~3 ppb in Benelux and UK. It is mainly due to the reduced NO_x emissions leading to decreased ozone titration (Aksoyoglu et al., 2014). The temporal analysis indicated that the increase in O_3 occured mostly in cold season and during the night when the O_3 mixing ratio was relatively low, and the peak O_3 in warm season actually decreased. Over the 2278 tested stations covering rural, urban and suburban sites in Europe, the maximum daily 8-hour mean ozone concentrations in summer (June–July–August) were ~2 ppb lower in 2020 compared to 2010.

The modelled concentration of NH_3 also increased in Europe except in Belarus although the NH_3 emissions were reduced by 6% in EU compared to the 2005 level. Similar increasing trends of NH_3 have already been observed in Europe from 2002 to 2016 (Warner et al., 2017). Because of the reduced SO_2 and NO_2 emissions, less NH_3 reacts to generate secondary inorganic aerosols, leading to more NH_3 in the atmosphere.

Implications on air quality management in Europe

Apart from the slight increase in ozone and NH_3, $PM_{2.5}$ appears to be a more critical issue for further air quality management in Europe. Figure 2a shows the country-level $PM_{2.5}$ concentrations in 2010 and 2020. While the yearly average limit for fine particles in Europe (25 μg m^{-3}) are well met, nearly half of the (20 out of 42) studied countries had average $PM_{2.5}$ exceeding the WHO AQG (10 μg m^{-3}) in 2010. The situation is improved by the reduced emissions in 2020, however, there are still 26% of the countries (11 out of 42) having average $PM_{2.5}$ above 10 μg m^{-3}. The local scale $PM_{2.5}$ level could be even higher than the country level average. As shown in Fig. 2b and 2c, large area in central Europe will still be exposed to $PM_{2.5}$ above the WHO AQG in 2020, indicating a high health risk to a considerable population in Europe.

Meanwhile, recent studies found that the health impacts of PM significantly depend on its composition and sources (Lelieveld and Poschl, 2017). According to the measurements of oxidative potential (OP, an indicator of capacity to generate oxidative stress and impact human health), metals from vehicular brake wear and organic aerosols from biomass burning are the major contributors to OP in Europe, while the OP of the inorganic aerosols are negligible (Dällenbach et al., 2018). However, in this study, we found that the reduction of $PM_{2.5}$ levels mostly come from the decrease in inorganic aerosols, indicating current emission reduction policy may have limited effects on reducing the real health impacts of PM.

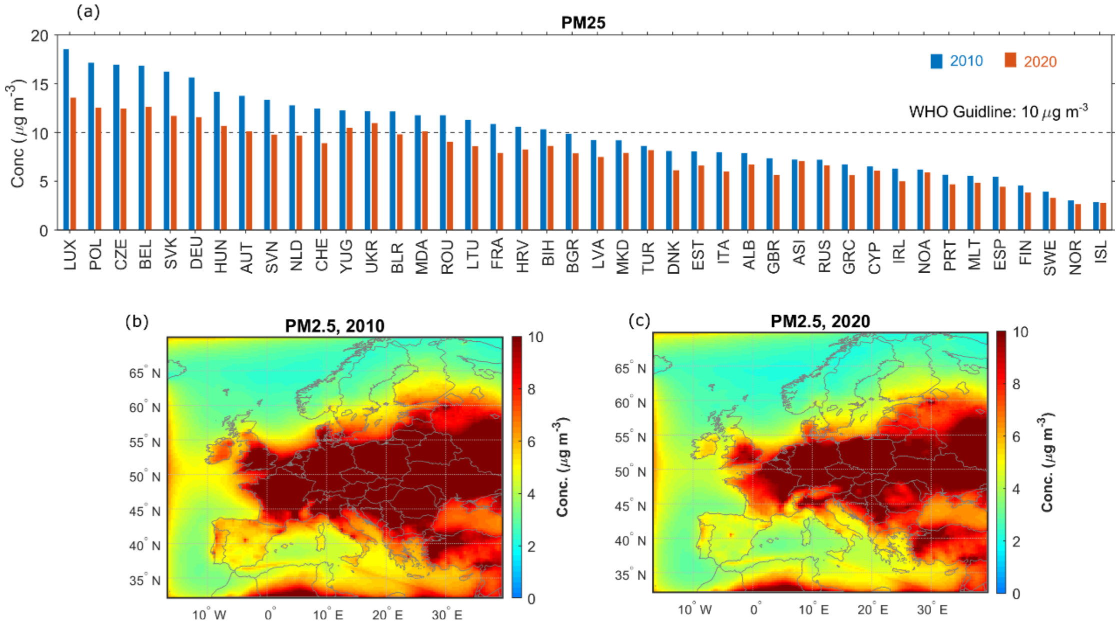

Figure 2. Average concentrations of $PM_{2.5}$ in country level between 2010 and 2020 (a) and spatial distribution of $PM_{2.5}$ concentrations in 2010 (b) and 2020 (c). The ISO Alpha-3 country codes are used. A full country list can be found in (Wikipedia, 2019). The dark red in the maps indicate grids value above the WHO AQG 10 µg m^{-3}.

CONCLUSION

This study investigated the effects of the revised Gothenburg Protocol on the European air quality by modelling 2010 and 2020 using the chemical transport model CAMx version 6.50. The results showed that the reduced emissions lead to a significant decline in the concentrations of the major pollutants SO_2, NO_x and $PM_{2.5}$ in 2020, while ozone and NH_3 levels slightly increase due to decreased O_3 titration, and reduced NO_x and SO_2 leading to a decrease in the formation of inorganic aerosols, respectively. Despite the predicted decrease in $PM_{2.5}$ levels, considerable population will still be exposed to high $PM_{2.5}$ exceeding the WHO air quality guidelines in 2020. The effects of decreased $PM_{2.5}$ concentrations on reducing health impacts remain to be evaluated in the context of recent findings showing that different PM components and sources have distinct health impacts.

ACKNOWLEDGMENTS

We thank Ramboll for support in CAMx modelling, EURODELTA-Trends project for providing meteorology, anthropogenic emission and boundary condition data for model input. This project was funded by the Swiss Federal Office of Environment (FOEN).

REFERENCES

Aksoyoglu, S., Keller, J., Ciarelli, G., Prevot, A. S. H., and Baltensperger, U., 2014: A model study on changes of European and Swiss particulate matter, ozone and nitrogen deposition between 1990 and 2020 due to the revised Gothenburg protocol, *Atmos. Chem. Phys.*, **14**, 13081-13095

Bessagnet, B., Pirovano, G., Mircea, M., Cuvelier, C., Aulinger, A., Calori, G., Ciarelli, G., Manders, A., Stern, R., Tsyro, S., Vivanco, M. G., Thunis, P., Pay, M. T., Colette, A., Couvidat, F., Meleux, F., Rouil, L., Ung, A., Aksoyoglu, S., Baldasano, J. M., Bieser, J., Briganti, G., Cappelletti, A., D'Isidoro, M., Finardi, S., Kranenburg, R., Silibello, C., Carnevale, C., Aas, W., Dupont, J. C., Fagerli, H., Gonzalez, L., Menut, L., Prévôt, A. S. H., Roberts, P., and White, L., 2016: Presentation of the EURODELTA III intercomparison exercise - evaluation of the chemistry transport models' performance on criteria pollutants and joint analysis with meteorology, *Atmos. Chem. Phys.*, **16**, 12667-12701

Boylan, J. W., and Russell, A. G., 2006: PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models, *Atmos. Environ.*, **40**, 4946-4959

Ciarelli, G., Aksoyoglu, S., Crippa, M., Jimenez, J. L., Nemitz, E., Sellegri, K., Äijälä, M., Carbone, S., Mohr, C., O'Dowd, C., Poulain, L., Baltensperger, U., and Prévôt, A. S. H., 2016: Evaluation of European air quality modelled by CAMx including the volatility basis set scheme, *Atmos. Chem. Phys.*, **2016**, 10313-10332

Colette, A., Andersson, C., Manders, A., Mar, K., Mircea, M., Pay, M. T., Raffort, V., Tsyro, S., Cuvelier, C., Adani, M., Bessagnet, B., Bergström, R., Briganti, G., Butler, T., Cappelletti, A., Couvidat, F., D'Isidoro, M., Doumbia, T., Fagerli, H., Granier, C., Heyes, C., Klimont, Z., Ojha, N., Otero, N., Schaap, M., Sindelarova, K., Stegehuis, A. I., Roustan, Y., Vautard, R., van Meijgaard, E., Vivanco, M. G., and Wind, P., 2017: EURODELTA-Trends, a multi-model experiment of air quality hindcast in Europe over 1990–2010, *Geosci. Model Dev.*, **10**, 3255-3276

Dällenbach, K. R., Uzu, G., Kourtchev, I., Cassagnes, L.-E., Vogel, A. L., Stefenelli, G., Vlachou, A., Slowik, J. G., Jaffrezo, J.-L., Kalberer, M., Dommen, J., Baltensperger, U., Haddad, I. E., and Prévôt, A. S. H., 2018: Relationship between aerosol composition and sources and their oxidative potential in central Europe, 10th International Aerosol Conference (IAC), St. Louis.

EEA, 2018: Air quality in Europe - 2018 report, European Environment Agency, Luxembourg.

EPA, 2007: Guidance on the use of models and other analyses for demonstrating attainment of air quality goals for ozone, PM2:5, and regional haze, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, North Carolina.

Guerreiro, C. B. B., Foltescu, V., and de Leeuw, F., 2014: Air quality status and trends in Europe, *Atmos. Environ.*, **98**, 376-384

Lelieveld, J., and Poschl, U., 2017: Chemists can help to solve the air-pollution health crisis, *Nature*, **551**, 291-293

Mol, W., and Leeuw, F., 2005: AirBase: a valuable tool in air quality assessments, Proceedings of the 5th International Conference on Urban Air Quality, Valencia, Spain.

Ramboll, 2018: User's guide: the Comprehensive Air Quality Model with extensions (CAMx) version 6.5, California.

UNECE, 2014: Guidance documents and other methodological materials for the implementation of the 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (Gothenburg Protocol): http://www.unece.org/environmental-policy/conventions/air/guidance-documents-and-other-methodological-materials/gothenburg-protocol.html.

Warner, J. X., Dickerson, R. R., Wei, Z., Strow, L. L., Wang, Y., and Liang, Q., 2017: Increased atmospheric ammonia over the world's major agricultural areas detected from space, *Geophys. Res. Lett.*, **44**, 2875-2884

Wikipedia, 2019: ISO 3166-1 alpha-3: https://en.wikipedia.org/w/index.php?title=ISO_3166-1_alpha-3&oldid=888744965.