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Effects of using two different biogenic emission models on ozone and particles in Europe

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Abstract

In this paper, we discuss the importance of biogenic volatile organic compound (BVOC) emissions used in air quality simulations and how the model results are affected by the choice of the BVOC emission model. The European air quality in 2011 was simulated using CAMx regional air quality model with two different BVOC emission models: PSI-model and MEGAN. Especially isoprene and monoterpene emissions calculated by the two models differed significantly both in amounts and their spatial distribution. In general, MEGAN produced much higher isoprene emissions while PSI-model generated more monoterpene emissions. The difference in emissions between the two models was shown to be as high as a factor of 3 in summer. The choice of the BVOC emission model had significant consequences especially on the formation of organic aerosols as well as on ozone and inorganic aerosols. Using MEGAN led to relatively higher ozone concentrations in summer while much more SOA (secondary organic aerosol) was formed when PSI-model was applied. Our results suggest that the amount and spatial distribution of BVOC emissions might affect the oxidant concentrations (OH and nitrate radicals, ozone) leading to significant differences in SOA, ozone, particulate nitrate and sulfate concentrations calculated by different BVOC emission models.

1 Introduction

The BVOC (biogenic volatile organic compounds) emissions have a significant influence on atmospheric reactions leading to formation of secondary pollutants such as ozone and secondary particles in many areas, especially in summer. In many inter-comparison studies, modelers share the same anthropogenic emissions, but the biogenic emissions usually differ (Bessagnet *et al.*, 2016), making the comparison of results from different models difficult. Since the uncertainties in BVOC emission estimates are very high (Simpson *et al.*, 2012; Oderbolz *et al.*, 2013), it is important to know the range of variability in simulated pollutant concentrations while using different BVOC emissions. In this study, we investigated the effects of using two different BVOC emission models on ozone as well as inorganic and organic particles in Europe during summer and winter periods in 2011.

2 Methods

We simulated the European air quality in 2011 using the regional air quality model CAMx (Comprehensive Air quality Model with extensions, v6.30, www.camx.com) and the meteorological model WRF-ARW (Weather Research and Forecasting Model, v3.7.1; Skamarock *et al.,* 2008). Our model domain covered Europe with a horizontal resolution of $0.25^o \times 0.125^o$ and there were fourteen terrain-following layers. The gas-phase mechanism was CB6r2 (Hildebrandt Ruiz and Yarwood, 2013). We used the fine/coarse option to calculate the concentrations of fine particles ($PM_{2.5}$) and the VBS scheme for organic aerosols. Initial and boundary conditions were obtained from the global model MOZART (Horowitz *et al.,* 2003). Anthropogenic emissions were based on the TNO-MACC-III inventory (Kuenen *et al.,* 2014). Two different biogenic emission models were used in this study to estimate BVOC emissions (isoprene, monoterpenes and sesquiterpenes) as well as NO emissions from soil. The first model was the PSI-model which was first developed by Andreani-Aksoyoglu and Keller (1995) and updated later by Oderbolz *et al.* (2013). It calculates BVOC emissions using temperature and photosynthetically active radiation (PAR) from WRF, land cover data from USGS and the vegetation inventory GlobCover (http://due.esrin.esa.int/page_globcover.php). The second model was the widely used MEGAN v2.1 (Guenther *et al.,* 2012) that covers 147 individual compounds within 19 categories. MEGAN also used the same meteorological data from WRF. The land use data was the Community Land Model version 4 (CLM4) and the leaf area index (LAI) was used to simulate changes of vegetation during the year. The ozone and organic aerosol (OA) concentrations calculated in two simulations using PSI-model and MEGAN, respectively, were compared with ACSM/AMS (Aerosol Chemical Speciation Monitor/Aerosol Mass Spectrometer) measurements at 8 European sites. Modelled ozone concentrations were compared with measurements at 537 rural AIRBASE (European Air Quality Database v7) stations.

3 Results and Discussion

We focussed on summer since the emissions in winter were much lower, especially for isoprene. The comparison of biogenic emissions from the two BVOC models suggests that MEGAN model generates more isoprene, but much less monoterpene emissions than the PSI-model in Europe (Figure 1) while the difference in sesquiterpene emissions was relatively small (<5%). In spite of three-times higher isoprene emissions in MEGAN, summer ozone was only slightly higher (<10%) than ozone calculated by the PSI-model (Figure 2, left panel). On the other hand, higher monoterpene emissions in the PSI-model led to higher SOA (Figure 2, right panel). Comparison of model results with measurements in Europe indicated that the bias for summer afternoon ozone mixing ratios higher than 50 ppb was lower when BVOC emissions were calculated with MEGAN, especially in southern Europe. For mixing ratios lower than 50 ppb however, the PSI-model showed a better performance. Mean bias between measured and modelled total organic aerosol was 8% - 90% lower by PSI model compared to MEGAN. Differences between the results of two simulations with different BVOC models were not only in ozone and SOA but also in particulate nitrate and sulfate, suggesting that the oxidant concentrations available for the formation of secondary inorganic aerosols might be affected significantly by the BVOC emissions, as also shown in Aksoyoglu *et al.* (2017).

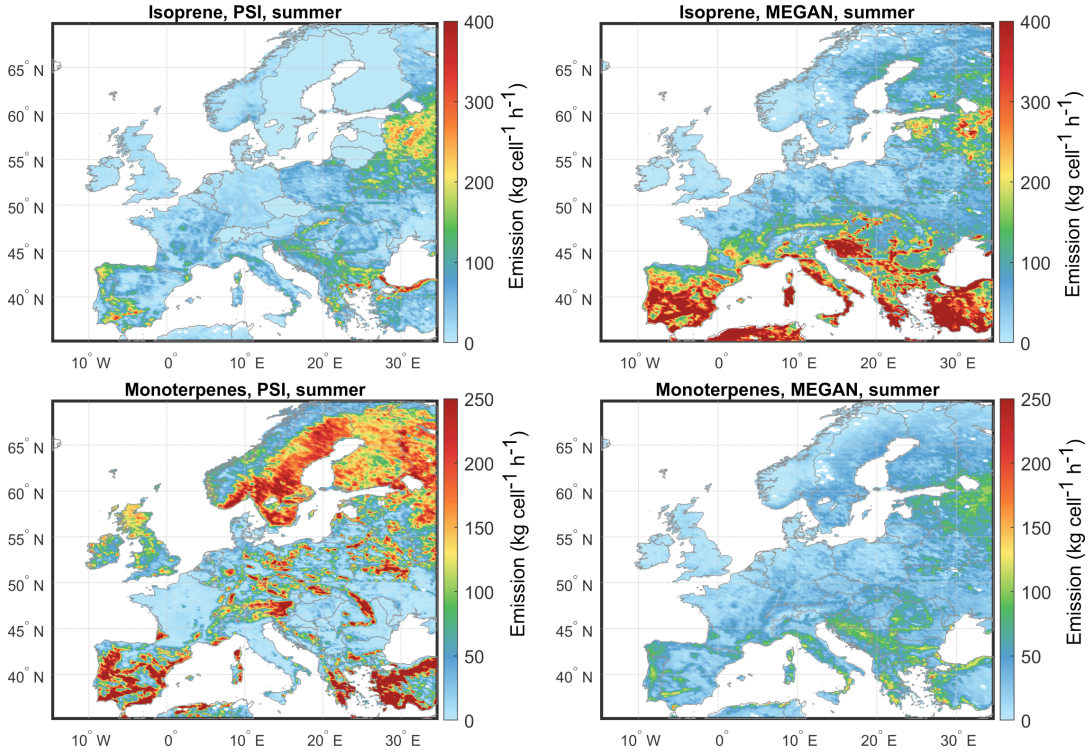

Figure 1. Isoprene (upper panels) and monoterpene (lower panels) emissions (kg cell^{-1} hr^{-1}) in July 2011 estimated by PSI-Model (left) and MEGAN (right).

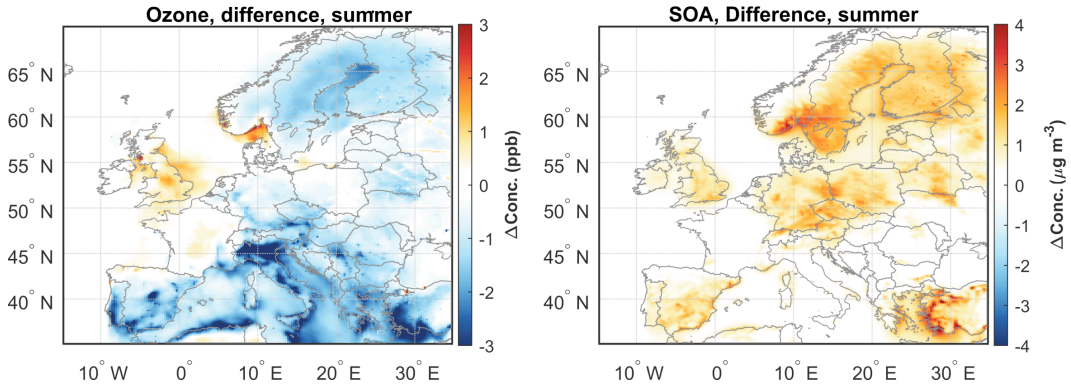

Figure 2. Difference in ozone (ppb) (left) and in SOA (µg m^{-3}) (right) in July 2011 between PSI-Model and MEGAN (PSI-model – MEGAN).

3 Conclusion

In this study, the European air quality in 2011 was simulated by CAMx using two biogenic volatile organic compound (BVOC) emission models: MEGAN and PSI-model. The results

showed that MEGAN generates more isoprene, but much less monoterpene emissions than the PSI-model in Europe probably due to their different land use and vegetation cover. In spite of much higher isoprene emissions generated by MEGAN compared to PSI-model, difference in ozone was relatively small (<10%) while three times higher monoterpene emissions in the PSI-model generated significantly more SOA (~110%) in summer. Comparison with measurements suggested a better performance with the PSI-model for organic aerosols while MEGAN showed a better agreement with measurements for high ozone levels. Particulate nitrate and sulfate concentrations were also affected by the BVOC emission model used. The results of this study emphasize the importance of BVOC emissions in air quality simulations and model inter-comparison studies.

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References

Andreani-Aksoyoglu, S., and Keller, J. (1995), "Estimates of monoterpene and isoprene emissions from the forests in Switzerland", *Journal of Atmospheric Chemistry,* Vol. 20, pp. 71-87, https://doi.org/10.1007/BF01099919.

Aksoyoglu, S., Ciarelli, G., El-Haddad, I., Baltensperger, U., and Prévôt, A. S. H. (2017), "Secondary inorganic aerosols in Europe: sources and the significant influence of biogenic VOC emissions, especially on ammonium nitrate", *Atmos. Chem. Phys.*, 17, 7757-7773, 10.5194/acp-17-7757-2017.

Bessagnet, B., Pirovano, G., Mircea, M., Cuvelier, C., Aulinger, A., Calori, G., Ciarelli, G., Manders, A., Stern, R., Tsyro, S., García Vivanco, M., Thunis, P., Pay, M. T., Colette, A., Couvidat, F., Meleux, F., Rouïl, 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.,* Vol. 16, pp. 12667-12701, 10.5194/acp-16-12667-2016.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., andWang, X. (2012), "The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions", *Geosci. Model Dev.,* Vol. 5, pp. 1471–1492, doi:10.5194/gmd 5-1471-2012.

Hildebrandt Ruiz, L. H., and Yarwood, G. (2013), Interactions between Organic Aerosol and NOy : Influence on Oxidant Production, Final Report for AQRP project 12-012.

Horowitz, L. W., Walters, S., Mauzerall, D. L., Emmons, L. K., Rasch, P. J., Granier, C., Tie, X., Lamarque, J. F., Schultz, M. G., and Tyndall, G. S. (2003), "A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2", *J. Geophys. Res.,* Vol. 108, pp. 4784.

Kuenen, J. J. P., Visschedijk, A. J. H., Jozwicka, M., and 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 modeling", *Atmos. Chem. Phys.,* Vol. 14, pp. 10963–10976, https://doi.org/10.5194/acp-14-10963-2014.

Oderbolz, D. C., Aksoyoglu, S., Keller, J., Barmpadimos, I., Steinbrecher, R., Skjøth, C. A., Plaß-Dülmer, C., and Prévôt, A. S. H. (2013), "A comprehensive emission inventory of biogenic volatile organic compounds in Europe: improved seasonality and land-cover", *Atmos. Chem. Phys.,* Vol.13, pp. 1689-1712, 10.5194/acp-13-1689-2013.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J. P., Valdebenito, Á., and Wind, P. (2012), "The EMEP MSC-W chemical transport model-technical description", *Atmos. Chem. Phys.,* Vol. 12, pp. 7825-7865, 10.5194/acp-12-7825-2012.

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X.-Y., Wang, W., and Powers, J. G. (2008), "A Description of the Advanced Research WRF Version 3", Mesoscale and Microscale Meteorology Division, National Center for Atmospheric Research, Boulder, Colorado, USA.

Questions and Answers

Questioner: Rostislav Kouznetsov
Question: Why do the state borders appear quite clearly in the BVOC emission maps of your model? It is especially well seen in isoprene emission maps from Scandinavia and Baltic states.
Answer: This is the case only for isoprene and it comes from using country-specific forest fractions, historically based on data from Simpson et al. (1999). In PSI-model the main isoprene emitting tree species is oak (a small fraction also emitted by Norway spruce). According to the country-specific data used, the isoprene-relevant forest fractions are zero in Sweden and Baltic countries, leading to clear borders in isoprene emission maps. This can be avoided in future by using other types of forest data.

Questioner: Vanisa Surapipith
Question: Is there any plan to improve PSI model? In particular, it is our intention to query for a possibility to do any necessary field campaign to improve the input for the biogenic VOC emission estimation. There is an interest to explore further this issue in our research institute in Thailand, there are very few studies so far in southeast Asia.
Answer: No, there is no such plan at the moment. PSI-model was developed originally only for Switzerland on a high-resolution domain using Swiss tree inventories and later it was extended to cover Europe.