Meteorology and Ozone, Temperature

J. Coates[1] and T. Butler[1]

[1]Institute for Advanced Sustainability Studies, Potsdam, Germany

September 1, 2015

Abstract

1 Introduction

2 Methodology

2.1 Model Setup

- MECCA box model as described in Coates and Butler (2015) to broadly simulate the Benelux (Belgium, Netherlands and Luxembourg) region. Solar zenith angle of 51°N was used to determine photolysis rates through a parameterisation and the SZA chosen is broadly representative of the central Benelux region.

- MECCA box model has been updated to include vertical mixing with the free troposphere and accordingly includes a diurnal cycle for the PBL height. These amendments are discussed further in Sect. 2.3.

- Simulations start at 06:00 using spring equinoctical conditions and the simulations ended after two days.

- All simulations performed using the Master Chemical Mechanism, MCM v3.2, (Rickard et al., 2015) and also repeated using MOZART-4 (Emmons et al., 2010). Coates and Butler (2015) describes the implementation of both MCM v3.2 and MOZART-4 for use with KPP within MECCA.

- NOx and other parameters were varied systematically to analyse the effects on ozone mixing ratios over different NOx gradients and hence different atmospheric conditions.

- VOC emissions constant until noon of first day, to simulate a plume of emitted VOC.

2.2 VOC Emissions

- Anthropogenic emissions from Benelux were obtained from the TNO-MACC_III emission inventory. TNO-MACC_III is the current version of the TNO-MACC_II inventory and was created using the same methodology as (Kuenen et al., 2014) and based upon improvements to the existing emission inventory during the AQMEII 2 exercies described in Pouliot et al. (2015).

- Temperature independent emissions of the biogenic VOC isoprene and monoterpenes, were calculated as a fraction of the total anthropogenic VOC emissions from each country in the Benelux region, this data was obtained from the supplementary data available from the EMEP (European Monitoring and Evaluation Programme) model (Simpson et al., 2012).

- AVOC and BVOC emissions are included as total emissions from SNAP (Selected Nomenclature for Air Pollution) source categories and these emissions were assigned to chemical groupings based on the country specific profiles for Belgium, the Netherlands and Luxembourg provided by TNO.

- The MCM v3.2 initial species were determined using the country specific profiles for each SNAP source category and where appropriate information of individual chemical species that can be represented by MCM v3.2 were determined using the speciations of Passant (2002).

- After calculating the MCM v3.2 initial VOC and respective emissions were assigned to the respective MOZART-4 species and the emissions in MOZART-4 were weighted by the carbon numbers of the MCM v3.2 species and the emitted MOZART-4 species.

Table 1: Anthropogenic NMVOC emissions in 2011 in tonnes from each SNAP category assigned from TNO-MACC_III emission inventory and biogenic VOC emission in tonnes from Benelux region assigned from EMEP. The allocation of these emissions to each MCM v3.2 and MOZART-4 species is found in the supplement.

	SNAP1	**SNAP2**	**SNAP34**	**SNAP5**	**SNAP6**	**SNAP71**
Belgium	4494	9034	22152	5549	42809	6592
Netherlands	9140	12173	29177	8723	53535	16589
Luxembourg	121	44	0	1372	4482	1740
Total	13755	21251	51329	15644	100826	24921
	SNAP72	**SNAP73**	**SNAP74**	**SNAP8**	**SNAP9**	**BVOC**
Belgium	2446	144	210	6449	821	6533
Netherlands	3230	1283	1793	10067	521	1356
Luxembourg	1051	6	324	643	0	2057
Total	6727	1433	2327	17159	1342	9946

Table 2: Belgium AVOC and BVOC emissions, in molecules cm^{-2} s^{-1}, translated into MCM species.

Type	MCM.species	SNAP.1	SNAP.2	SNAP.34	SNAP.5	SNAP.6	SNAP.71	SNAP.72	SNAP.73	SNAP.74	SNAP.8	SNAP.9	BVOC	Total
Ethane	C2H6	4.15E+08	1.11E+09	2.98E+09			1.74E+08	4.62E+07	8.17E+06		8.30E+07	8.22E+07		4.91E+09
Propane	C3H8	1.14E+09	4.72E+08	1.03E+08	3.12E+10	3.18E+08	8.49E+06	3.15E+07	8.17E+07	2.71E+06	7.53E+07	3.56E+07		3.35E+10
Butanes	NC4H10	7.77E+08	2.42E+08	1.27E+06	1.23E+11	1.18E+09	1.89E+08	3.26E+07		4.48E+07	1.40E+08	2.20E+07		1.25E+11
	IC4H10	9.48E+07	8.49E+07	3.11E+05	2.98E+10	5.36E+07	8.81E+07	1.52E+07		2.09E+07	7.02E+07	2.20E+07		3.03E+10
Pentanes	NC5H12	6.21E+08	2.25E+08		8.78E+10		1.13E+08	1.31E+07		2.25E+07	4.51E+07	1.11E+07		8.89E+10
	IC5H12	2.62E+08	1.21E+08		5.25E+10		2.19E+08	2.54E+07		4.37E+07	8.60E+07	1.11E+07		5.33E+10
	NEOP											1.11E+07		1.11E+07
Hexane and Higher Alkanes	NC6H14	3.89E+08	2.39E+07	3.15E+08	1.26E+10	1.05E+09	3.98E+08	1.94E+08		8.35E+06	1.04E+08	3.84E+06		1.51E+10
	M2PE			4.06E+07	1.94E+09	2.20E+08					1.73E+08	1.65E+06		2.37E+09
	M3PE			3.04E+07	9.69E+08	2.20E+08					1.04E+08			1.32E+09
	NC7H16	1.67E+08	4.11E+07	1.48E+08	1.35E+10	3.79E+08	6.55E+07	3.20E+07		1.38E+06	2.98E+07	1.94E+07		1.44E+10
	M2HEX					1.42E+08	5.10E+07	2.49E+07		1.07E+06	4.48E+07			2.64E+08
	M3HEX					1.42E+08	3.64E+07	1.78E+07		7.64E+05	2.98E+07			2.27E+08
	M22C4										3.47E+07			3.47E+07
	M23C4										3.47E+07			3.47E+07
	NC8H18			6.13E+07	1.01E+10	4.16E+07	5.75E+07	2.81E+07		1.21E+06	1.70E+08	6.63E+06		1.04E+10
	NC9H20			3.41E+07		1.00E+09						2.21E+06		1.04E+09
	NC10H22			4.30E+07		1.94E+09	2.56E+07	1.25E+07		5.38E+05		3.32E+06		2.02E+09
	NC11H24			1.68E+07		7.90E+08	9.33E+06	4.56E+06		1.96E+05	1.91E+07	1.21E+06		8.41E+08
	NC12H26					5.58E+07	1.52E+08	7.44E+07		3.20E+06	1.76E+07			3.03E+08
	CHEX		3.81E+07	1.04E+07		2.26E+08						1.12E+06		2.75E+08
Ethene	C2H4	8.93E+07	2.49E+09	3.11E+10			9.61E+08	5.94E+08	4.38E+07		1.18E+09	1.43E+08		3.66E+10
Propene	C3H6	5.95E+07	5.21E+08	5.33E+08			3.38E+08	9.90E+07	1.95E+07		2.06E+08	4.10E+07		1.82E+09
Higher Alkenes	HEX1ENE	5.05E+06	1.28E+07									1.63E+07		3.42E+07
	BUT1ENE		1.80E+07	6.24E+07							1.96E+07			9.99E+07
	MEPROPENE										9.80E+06			9.80E+06
	TBUT2ENE										9.80E+06			9.80E+06
	CBUT2ENE										9.80E+06			9.80E+06
	CPENT2ENE		5.65E+06								3.92E+06			9.57E+06
	TPENT2ENE		5.65E+06								3.92E+06			9.57E+06
	PENT1ENE		5.14E+06	5.93E+06							1.57E+07			2.68E+07
	ME2BUT2ENE		3.08E+06								7.84E+06			1.09E+07
	ME3BUT1ENE		3.08E+06								7.84E+06			1.09E+07
	ME2BUT1ENE		2.05E+06											2.05E+06
Ethyne	C2H2	6.97E+05	7.84E+08	3.45E+08			8.95E+08	2.80E+08	1.73E+07	1.09E+07	3.95E+08	5.38E+07		2.78E+09

Type	MCM.species	SNAP.1	SNAP.2	SNAP.34	SNAP.5	SNAP.6	SNAP.71	SNAP.72	SNAP.73	SNAP.74	SNAP.8	SNAP.9	BVOC	Total
Benzene	BENZENE	6.91E+07	4.64E+08	5.74E+08	3.05E+09		2.16E+08	3.56E+07		1.53E+06	7.98E+07	2.75E+07		4.52E+09
Toluene	TOLUENE	8.49E+07	1.54E+08	4.87E+07	2.59E+09	2.16E+09	4.88E+08	2.26E+07		1.30E+06	6.79E+07	1.81E+07		5.63E+09
Xylenes	MXYL	4.20E+07	1.32E+07	1.60E+06	3.74E+08	1.25E+09	1.04E+08	9.52E+06		2.05E+05	1.86E+07	3.66E+06		1.81E+09
	OXYL	9.33E+06	1.32E+07	6.42E+05	3.74E+08	3.12E+08	1.04E+08	9.52E+06		2.05E+05	1.51E+07	2.19E+06		8.40E+08
	PXYL		1.32E+07	6.42E+05	3.74E+08	3.12E+08	7.79E+07	7.14E+06		1.53E+05	1.86E+07	2.93E+06		8.07E+08
Trimethylbenzenes	TM123B	6.21E+03	1.06E+06			2.09E+07	1.79E+07				3.33E+06	3.30E+05		4.35E+07
	TM124B	6.21E+03	1.06E+06	1.46E+07		7.11E+07	7.50E+07				7.76E+06	4.40E+05		1.70E+08
	TM135B	6.21E+03	1.06E+06			2.09E+07	2.86E+07				3.33E+06	4.40E+05		5.43E+07
Other Aromatics	EBENZ	1.36E+07		1.65E+07		5.68E+07	7.76E+07	5.32E+07	1.53E+04		1.74E+08	3.93E+06		3.96E+08
	PBENZ					1.26E+07	6.86E+07	4.70E+07	1.35E+04		2.79E+07	1.73E+06		1.58E+08
	IPBENZ					4.60E+07					2.79E+07	1.73E+06		7.57E+07
	PETHTOL					4.18E+06					5.59E+07			6.00E+07
	METHTOL					1.26E+07					5.59E+07			6.84E+07
	OETHTOL										4.19E+07			4.19E+07
	DIET35TOL						1.45E+08	9.94E+07	2.86E+04					2.45E+08
	DIME35EB					7.12E+07	1.79E+07	1.23E+07	3.53E+03					1.01E+08
	STYRENE			1.68E+07		1.45E+07	1.65E+07	1.13E+07	3.25E+03					5.91E+07
	BENZAL						2.77E+07	1.90E+07	5.46E+03					4.68E+07
	PHENOL			1.86E+07										1.86E+07
Formaldehyde	HCHO	2.74E+07	5.76E+08				2.12E+08	2.78E+08	1.09E+07		1.23E+09	2.22E+07		2.35E+09
Other Aldehydes	CH3CHO	2.82E+06	7.80E+07	7.07E+07			5.74E+07	1.15E+08	2.09E+06		2.22E+08	5.17E+06		5.53E+08
	C2H5CHO	1.61E+06	5.91E+07				9.67E+06	1.94E+07	3.52E+05		8.41E+07	3.92E+06		1.78E+08
	C3H7CHO	1.29E+04	4.76E+07								6.78E+07	3.16E+06		1.19E+08
	IPRCHO	1.29E+04	4.76E+07								4.52E+07	3.16E+06		9.60E+07
	C4H9CHO	1.08E+04	3.99E+07									2.64E+06		4.25E+07
	ACR	1.67E+04	6.13E+07				1.50E+07	3.02E+07	5.48E+05			4.06E+06		1.11E+08
	MACR	1.33E+04	4.90E+07									3.25E+06		5.23E+07
	C4ALDB	1.33E+04	4.90E+07				8.01E+06	1.61E+07	2.92E+05			3.25E+06		7.67E+07
	MGLYOX										4.52E+07			4.52E+07
Alkadienes and	C4H6	1.32E+07	2.34E+08	3.10E+08	2.09E+10		4.51E+08	1.21E+08	3.14E+07	1.98E+07	2.84E+08	1.98E+07		2.24E+10
Other Alkynes	C5H8	1.05E+07	1.86E+08		1.66E+10							1.58E+07	3.11E+09	2.00E+10
Organic Acids	HCOOH	1.27E+06	7.07E+08								1.67E+08	5.23E+07		9.28E+08
	CH3CO2H	9.72E+05	5.42E+08	4.37E+07							1.28E+08	4.01E+07		7.55E+08
	PROPACID	7.88E+05	4.39E+08								1.04E+08	3.25E+07		5.77E+08
	ACO2H			3.64E+07										3.64E+07

Type	MCM.species	SNAP.1	SNAP.2	SNAP.34	SNAP.5	SNAP.6	SNAP.71	SNAP.72	SNAP.73	SNAP.74	SNAP.8	SNAP.9	BVOC	Total
Alcohols	CH3OH	5.18E+04		2.12E+06		2.00E+09					4.03E+07	1.81E+07		2.07E+09
	C2H5OH	3.60E+04	9.73E+08	5.98E+07		2.05E+09					2.80E+07	4.77E+07		3.16E+09
	NPROPOL	2.76E+04				1.67E+08					2.15E+07	5.78E+06		1.94E+08
	IPROPOL	2.76E+04		7.52E+05		2.67E+08					2.15E+07			2.89E+08
	NBUTOL	2.24E+04				1.62E+08					1.74E+07			1.80E+08
	BUT2OL	2.24E+04				1.08E+08					1.74E+07	7.80E+06		1.34E+08
	IBUTOL	2.24E+04				6.77E+07					1.74E+07			8.51E+07
	TBUTOL	2.24E+04									1.74E+07			1.74E+07
	PECOH	1.88E+04									1.46E+07			1.47E+07
	IPEAOH	1.88E+04									1.46E+07			1.47E+07
	ME3BUOL	1.88E+04									1.46E+07			1.47E+07
	IPECOH	1.88E+04									1.46E+07			1.47E+07
	IPEBOH	1.88E+04									1.46E+07			1.47E+07
	CYHEXOL	1.66E+04									1.29E+07			1.29E+07
	MIBKAOH	1.43E+04				3.46E+07					1.11E+07			4.57E+07
	ETHGLY	2.67E+04				4.85E+07					2.08E+07			6.93E+07
	PROPGLY	2.18E+04				9.67E+07					1.69E+07			1.14E+08
	C6H5CH2OH					2.78E+07								2.78E+07
	MBO	1.93E+04									1.50E+07			1.50E+07
Ketones	CH3COCH3	1.29E+05	1.08E+07	1.66E+08		2.13E+09	6.45E+06	3.59E+07			1.73E+08	1.06E+06		2.53E+09
	MEK		8.73E+06			1.03E+09						8.54E+05		1.04E+09
	MPRK		7.31E+06									7.15E+05		8.03E+06
	DIEK		7.31E+06									7.15E+05		8.03E+06
	MIPK		7.31E+06									7.15E+05		8.03E+06
	HEX2ONE		6.29E+06									6.15E+05		6.90E+06
	HEX3ONE		6.29E+06									6.15E+05		6.90E+06
	MIBK		6.29E+06			6.18E+08						6.15E+05		6.25E+08
	MTBK		6.29E+06									6.15E+05		6.90E+06
	CYHEXONE		6.42E+06	8.91E+06		5.05E+07						6.28E+05		6.64E+07
Terpenes	APINENE											2.28E+06	3.89E+08	3.91E+08
	BPINENE											2.28E+06	3.89E+08	3.91E+08
	LIMONENE					6.87E+07						3.42E+06	3.89E+08	4.61E+08
Esters	METHACET			6.18E+07										6.18E+07
	ETHACET			7.08E+06		1.38E+09								1.39E+09
	NBUTACET					9.65E+08								9.65E+08
	IPROACET					3.40E+08								3.40E+08
	CH3OCHO			6.93E+06										6.93E+06
	NPROACET					1.27E+08						5.94E+06		1.33E+08

Type	MCM.species	SNAP.1	SNAP.2	SNAP.34	SNAP.5	SNAP.6	SNAP.71	SNAP.72	SNAP.73	SNAP.74	SNAP.8	SNAP.9	BVOC	Total
Ethers	CH3OCH3		3.36E+07	2.43E+08		7.77E+07								3.54E+08
	DIETETHER		2.09E+07	9.06E+07										1.11E+08
	MTBE		1.76E+07											1.76E+07
	DIIPRETHER		1.52E+07	6.57E+07								1.47E+07		9.56E+07
	ETBE		1.52E+07											1.52E+07
	MO2EOL		2.04E+07			9.40E+07								1.14E+08
	EOX2EOL		1.72E+07			7.94E+07								9.66E+07
	PR2OHMOX		1.72E+07			1.59E+08								1.76E+08
	BUOX2ETOH		1.31E+07			7.62E+08								7.75E+08
	BOX2PROL		1.17E+07											1.17E+07
Chlorinated Hydrocarbons	CH2CL2			1.75E+08		6.16E+08						1.09E+06		7.92E+08
	CH3CH2CL			1.36E+08										1.36E+08
	CH3CCL3					4.31E+08						3.47E+05		4.31E+08
	TRICLETH			6.66E+07		9.75E+08						3.52E+05		1.04E+09
	CDICLETH			4.51E+07								7.11E+05		4.58E+07
	TDICLETH			4.51E+07								4.74E+05		4.56E+07
	CH3CL			1.39E+08										1.39E+08
	CCL2CH2			4.51E+07										4.51E+07
	CHCL2CH3											5.35E+05		5.35E+05
	VINCL			4.20E+07										4.20E+07
	TCE			1.05E+07		2.36E+08						6.93E+05		2.48E+08
	CHCL3			2.93E+07										2.93E+07
Total		4.30E+09	1.12E+10	3.85E+10	4.07E+11	2.73E+10	6.00E+09	2.47E+09	2.16E+08	1.85E+08	6.61E+09	8.82E+08	4.28E+09	5.09E+11

2.3 Vertical Mixing with Diurnal Boundary Layer Height

- The base boxmodel (Sect. 2.1) includes a constant boundary layer height of 1 km and no interactions (mixing) with the free troposphere.

- A parameterisation of the diurnal profile of the planetary boundary layer (PBL) height over Los Angeles was provided by Boris Bonn based on data from the CARES field campaign (CARB, 2008) . check if reference correctly

- The PBL height was calculated at every time point for the model run and then read into the boxmodel at each time point . include plot

- The concentrations of the chemical species within the PBL are diluted due to the larger mixing volume when the PBL height increases at the beginning of the day, also the increasing PBL height induces mixing of chemical species from the free troposphere with those chemical species within the PBL i.e. vertical mixing. When the PBL height collapses during night giving the stable nocturnal boundary layer, this traps the chemical species into a smaller volume thus increasing the concentrations of the chemical species.

- This vertical mixing scheme was implemented into the boxmodel using the same approach of Lourens (2012).

- The mixing ratios of O3, CO and CH4 in the free troposphere were respectively set to 50 ppbv, 116 ppbv and 1.8 ppmv. These condions were taken from the MATCH-MPIC chemical weather forecast model on the 27th March (the start date of the simulations). The model results (`http://cwf.iass-potsdam.de/`) at the 700 hPa height were chosen and the daily average was used as input into the boxmodel. check reference

- Tagged free troposphere species were also included in the boxmodel to determine effect of free troposphere species on surface ozone levels.

3 Results

4 Conclusions

References

CARB. 2010 CalNex White Paper: Research at the Nexus of Air Quality and Climate Change. Technical report, California Air Resources Board, 2008.

J. Coates and T. M. Butler. A comparison of chemical mechanisms using tagged ozone production potential (TOPP) analysis. *Atmospheric Chemistry and Physics*, 15(15):8795–8808, 2015.

L. K. Emmons, S. Walters, P. G. Hess, J.-F. Lamarque, G. G. Pfister, D. Fillmore, C. Granier, A. Guenther, D. Kinnison, T. Laepple, J. Orlando, X. Tie, G. Tyndall, C. Wiedinmyer, S. L. Baughcum, and S. Kloster. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). *Geoscientific Model Development*, 3:43–67, 2010.

J. J. P. Kuenen, A. J. H. Visschedijk, M. Jozwicka, and H. A. C. Denier van der Gon. TNO-MACC_II emission inventory; a multi-year (2003–2009) consistent high-resolution european emission inventory for air quality modelling. *Atmospheric Chemistry and Physics*, 14(20): 10963–10976, 2014.

AsM Lourens. *Air quality in the Johannesburg-Pretoria megacity: its regional influence and identification of parameters that could mitigate pollution.* PhD thesis, North-West University, Potchefstroom Campus, 2012.

N. Passant. Speciation of UK emissions of non-methane volatile organic compounds. Technical report, DEFRA, Oxon, UK., 2002.

George Pouliot, Hugo A.C. Denier van der Gon, Jeroen Kuenen, Junhua Zhang, Michael D. Moran, and Paul A. Makar. Analysis of the emission inventories and model-ready emission datasets of Europe and North America for phase 2 of the AQMEII project. *Atmospheric Environment*, 115: 345–360, 2015.

Andrew Rickard, Jenny Young, M. J. Pilling, M. E. Jenkin, Stephen Pascoe, and S. M. Saunders. The Master Chemical Mechanism Version MCM v3.2. `http://mcm.leeds.ac.uk/MCMv3.2/`, 2015. [Online; accessed 25-March-2015].

D. Simpson, A. Benedictow, H. Berge, R. Bergström, L. D. Emberson, H. Fagerli, C. R. Flechard, G. D. Hayman, M. Gauss, J. E. Jonson, M. E. Jenkin, A. Nyíri, C. Richter, V. S. Semeena, S. Tsyro, J.-P. Tuovinen, Á. Valdebenito, and P. Wind. The EMEP MSC-W chemical transport model – technical description. *Atmospheric Chemistry and Physics*, 12(16):7825–7865, 2012.