AEROSOL MODELING AT REGIONAL AND CONTINENTAL SCALES

Karine Sartelet*, Edouard Debry, Marilyne Tombette, Yelva Roustan, Bruno Sportisse
CEREA, Joint Research Laboratory, École Nationale des Ponts et Chaussées / EDF R&D, France

This talk aims at presenting two aerosol models that are part of the air-quality modeling system Polyphemus : SIREAM (SIze Resolved Aerosol Model) based on a sectional description of the aerosol distribution and MAM (Modal Aerosol Model) based on a modal discretization. The models are first described, as well as their coupling to the chemistry-transport model Polair3D of Polyphemus. Validations were made at the regional scale over Paris and Tokyo, and at the continental scales over Europe and over Asia in the framework of MICS (Model Inter-Comparison Study). Sensitivity of aerosol concentrations to configuration choices used in the aerosol models, such as whether or not the assumption of thermodynamic equilibrium is made for coarse aerosols, is briefly presented.

1 MODELING

The models SIREAM and MAM are described in details in Debry et al. [2007] and Sartelet et al. [2006]. SIREAM (SIze Resolved Aerosol Model) based on a sectional description of the aerosol distribution and MAM (Modal Aerosol Model) based on a modal discretization. The models share physical parameterizations. They both assume internal mixing of inert (mineral dust, elemental carbon), inorganic (sodium, chloride, nitrate, sulfate and ammonium) and organic species (primary organics and secondary organics following Schell et al. [2001]). The following processes are modeled : ternary nucleation following Napari et al. [2002], Brownian coagulation, condensation/evaporation solved using ISORROPIA (Nenes et al. [1998]). Mass transfer can be solved either assuming thermodynamic equilibrium (bulk equilibrium approach) or dynamically. In addition, a hybrid approach can be used in which thermodynamic equilibrium is assumed for the smallest sections/modes and mass transfer is computed dynamically for the largest sections/modes. In the framework of the air quality modeling system Polyphemus, SIREAM/MAM are coupled to the chemistry transport model Polair3D (Mallet et al. [2007]). Heterogeneous reactions : $HO_2 \rightarrow 0.5\ H_2O_2$, $NO_2 \rightarrow 0.5\ HONO + 0.5\ HNO_3$, $NO_3 \rightarrow HNO_3$, $N_2O_5 \rightarrow 2\ HNO_3$ are modeled following Jacob [2000] with the reaction probabilities $\gamma_{HO_2} = 0.1$, $\gamma_{NO_2} = 10^{-6}$, $\gamma_{NO_3} = 2.10^{-4}$, $\gamma_{N_2O_5} = 0.01$, except when specified. When the liquid water content of the cell exceeds a threshold value of 0.05g m^{-3}, the grid cell is assumed to contain a cloud, and a model for aqueous phase chemistry for cloud dropets (based on the Variable Size Resolved Model VSRM of Fahey and Pandis [2003]) is called instead of SIREAM/MAM.

*Corresponding author : sartelet@cerea.enpc.fr

2 VALIDATION AND SENSITIVITY STUDIES

The validation of Polyphemus-SIREAM or Polyphemus-MAM was done at the regional scale over Paris (Tombette and Sportisse [2007]), Greater Tokyo (Sartelet et al. [2007b]), at the regional scale over Asia (Sartelet et al. [2007c]) and over Europe (Sartelet et al. [2007a]). The domain and input data are not detailed here, and the comparisons to measurements are only briefly summarized. The scores obtained for PM_{10} and inorganic aerosols (sulfate and nitrate) are presented (measured and simulated mean, correlation, normalized mean error NME and normalized mean bias NMB, see Sartelet et al. [2007c] for a definition of these statistics). Different assumptions can be made in the modeling and different parameterizations can be used. To test the impact of aerosol processes and modeling assumptions on aerosol concentrations, sensitivity studies were carried out for each validation. The reference simulation is the simulation used to compute the scores. The reference simulation is compared to simulations where only one aerosol process differ from the reference simulation. The comparison is done by computing the NME between the reference simulation and the other simulations. Although the impact of aerosol processes on aerosol concentrations differ depending on local conditions and the chemical component studied, comparisons of the different sensitivity studies allow us to point out common features and differences.

2.1 At the regional scale

2.1.1 over Greater Paris

Over Greater Paris, Tombette and Sportisse [2007] run a simulation for 5 months, from 1 May 2001 to 30 September 2001, using 10 sections in the range 0.01μm - 10μm. The scores for PM_{10} averaged over 8 stations are given in table 1.

A sensitivity study was conducted over the first 15 days of July. All the discretization points in the domain of study are taken into account when computing the statistics. Three simulations are considered in the sensitivity study : a run without heterogeneous reaction (the reaction probabilities are ta-

TAB. 1 – Scores for PM_{10} over Greater Paris

Meas. mean	Sim. mean	Corr	NMB	NME
23.0	23.9	59.5	4.0	32.4

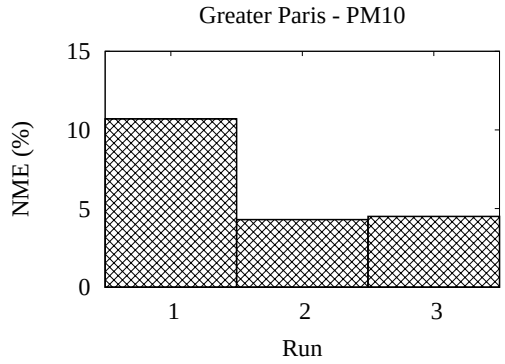

FIG. 1 – Over Greater Paris, for PM_{10}, differences between the reference run and a run without heterogeneous reaction (1), bulk equilibrium (2) and dynamic mass transfer (3)

ken as $\gamma_{HO_2} = 0.2$, $\gamma_{NO_2} = 10^{-5}$, $\gamma_{NO_3} = 10^{-3}$, $\gamma_{N_2O_5} = 0.03$ in the reference run), a run assuming bulk equilibrium for all sections instead of computing dynamically the largest 3 sections (i.e. sections of diameter larger than 1.25μm), and a run where all sections are computed dynamically. As shown in Figure 1, PM_{10} concentrations were found to be very sensitive to heterogeneous reaction with a NME of 11$\%$ but less sensitive to whether mass transfer is computed dynamically or assuming bulk equilibrium (the NME is about 4$\%$).

2.1.2 over Greater Tokyo

Over Greater Tokyo, Sartelet et al. [2007b] use Polyphemus-MAM to simulate two high-pollution episodes : one in winter (9 and 10 December 1999) and one in summer (31 July and 1 August 2001). The 4 modes lie in the diameter range 0.001μm and 10μm.

The scores are given in table 2 for inorganic $PM_{2.5}$ (sulfate and nitrate) at 4 stations for the winter episode and two stations for the summer episode. Better scores are generally observed for sulfate than nitrate.

Only the concentrations at the location of measurements were considered in the sensitivity study. In the reference run, condensation assuming thermodynamic equilibrium, dry deposition, nucleation and coagulation are taken into account, heterogeneous reactions are ignored. Eight simulations were conducted : without condensation, using the hybrid scheme (thermodynamic equilibrium is assumed only for the smallest mode instead of the four

TAB. 2 – Scores for sulfate and nitrate over Tokyo in the winter episode (first 2 lines) and in the summer episode (last 2 lines)

	Meas.	Sim.	Corr	NMB	NME
sulfate	2.6	3.3	66.2	-26	41
nitrate	4.5	4.1	44.5	8	111
sulfate	13.3	14.1	-2	-7	34
nitrate	5.7	2.1	34	62	67

modes), with heterogeneous reactions (the reaction probabilities are taken as $\gamma_{HO_2} = 0.2$, $\gamma_{NO_2} = 10^{-4}$, $\gamma_{NO_3} = 10^{-3}$, $\gamma_{N_2O_5} = 0.03$ in the reference run), without deposition, without nucleation, without coagulation, using SIREAM instead of MAM (i.e. using the sectional distribution instead of the modal distribution), using CMAQ instead of Polyphemus-MAM. Because CMAQ models the same processes as Polyphemus-MAM, comparison of the reference run to CMAQ allows us to have a rough idea of the sensitivity to parameterizations in and outside the aerosol module.

Comparisons of the NME between each of these simulations and the reference run is shown in Figure 2. For sulfate, in the summer episode, the impact of long-range transport dominates because the close by Mount Oyama of Miyake Island was in eruption at that time. This is illustrated by the fact that the highest sensitivity is observed when CMAQ is used (with a NME of 42$\%$ against 12$\%$ for condensation). In the winter episode, sulfate is mostly impacted by condensation (21$\%$), CMAQ (20$\%$), coagulation (17$\%$), and deposition to a lesser extent(10$\%$). Whereas nucleation and coagulation are negligible in the summer episode (1$\%$), they are not in the winter episode (17$\%$ and 5$\%$). The impact of coagulation is larger in the winter episode than in the summer episode, because the number of small particles is higher in the winter episode as a consequence of higher nucleation.

For nitrate, the impacts of condensation, heterogeneous reactions and CMAQ dominate (66$\%$, 190$\%$ and 100$\%$ respectively in the winter episode and 99$\%$, 119$\%$ and 251$\%$ in the summer episode). The impact of the thermodynamic equilibrium assumption is limited (12$\%$ in the winter episode and 49$\%$ in the summer episode).

For both sulfate and nitrate, the impact of dry deposition is limited (8 to 10$\%$).

The impact of using a sectional representation of the size distribution is small, also not negligible, and it is higher for nitrate (8-18$\%$) than for sulfate (6-8$\%$).

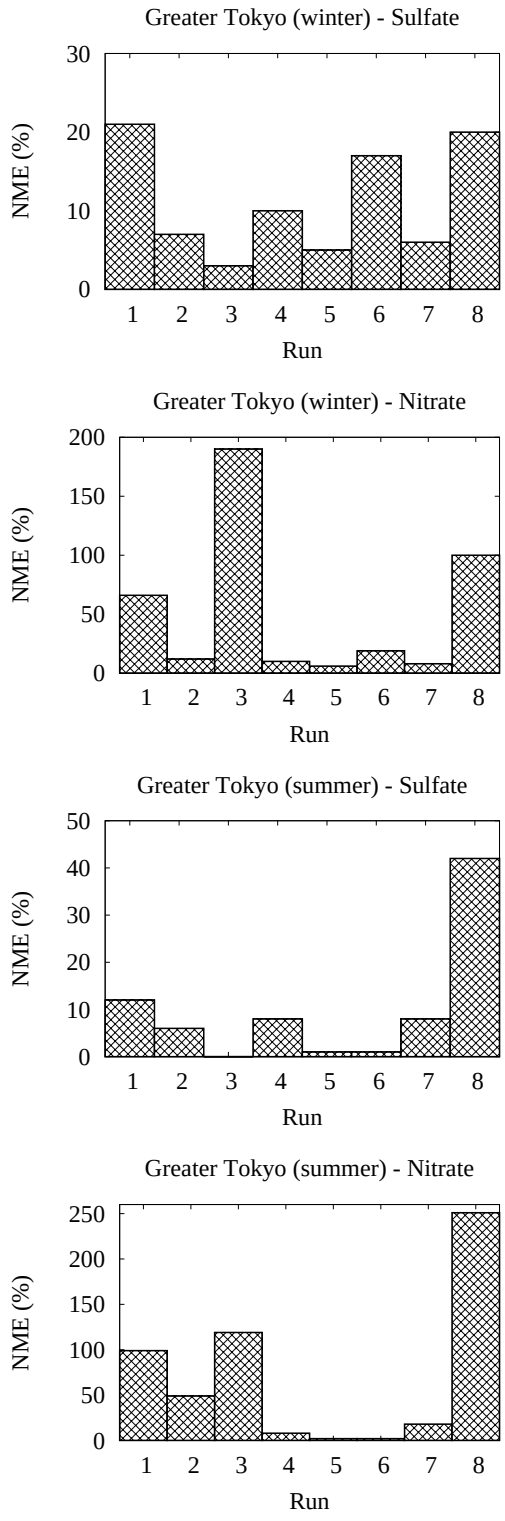

FIG. 2 – Over Greater Tokyo, for sulfate and nitrate, differences between the reference run and a run without condensation (1), using the hybrid scheme (2), with heterogeneous reaction (3), without dry deposition (4), without nucleation (5), without coagulation (6), SIREAM (7), CMAQ (8)

TAB. 3 – Scores for sulfate and nitrate over 24 stations of East Asia in March 2001

	Meas.	Sim.	NMB	NME
sulfate	4.9	5.2	13	13
nitrate	0.9	1.8	24	27

2.2 At the continental scale

2.2.1 over East Asia

The validation of Polyphemus-SIREAM over East Asia was done in the framework of the model intercomparison study–Asia Phase II (MICS2).The sensitivity study was conducted only for March 2001. The scores for sulfate and nitrate over 24 stations are given in table 3. In the reference run, 10 sections in the range 0.01μm - 10μm are used, thermodynamic equilibrium is assumed, heterogeneous reactions and cloud chemistry are ignored. Only the concentrations at the location of measurements were considered in the sensitivity study. Seven simulations were conducted : with 3 sections instead of 10, with the hybrid scheme (with a cutoff diameter of 0.6μm, i.e. the finest 6 sections are computed with the full equilibrium approach), without coagulation, with heterogeneous reaction ($\gamma_{HO_2} = 0.2$, $\gamma_{NO_2} = 10^{-4}$, $\gamma_{NO_3} = 10^{-3}$, $\gamma_{N_2O_5} = 0.03$), with cloud chemistry, using CMAQ instead of Polyphemus-SIREAM.

As shown in Figure 3, sulfate is mostly impacted by CMAQ (34$\%$), and by cloud chemistry (17$\%$), heterogeneous reaction (15$\%$) and coagulation (11$\%$) to a lesser extent. For nitrate, the impact of heterogeneous reactions is very large (99$\%$) followed by CMAQ (71$\%$). The number of sections, the hybrid scheme and coagulation have much smaller impacts although they are not negligeable (15$\%$, 13$\%$ and 15$\%$ respectively).

2.2.2 over Europe

Sartelet et al. [2007a] presented a validation of multi-pollutants over Europe with a focus on aerosols for the year 2001 (see the scores in table 4 evaluated with three different databases (EMEP, AirBase and BDQA). A sensitivity study was conducted over 1 month in summer (between 15 July and 14 August 2001) and 1 month in winter (between 15 November and 14 December 2001).

All the discretization points in the domain of study are considered when computing the statistics. In the reference run, 5 sections in the range 0.01μm - 10μm are used, thermodynamic equilibrium is assumed, heterogeneous reactions and cloud chemistry are taken into account, nucleation is ignored. Twelve other simulations were conducted : with 10 sections instead of 5, with dynamic mass transfer for all sec-

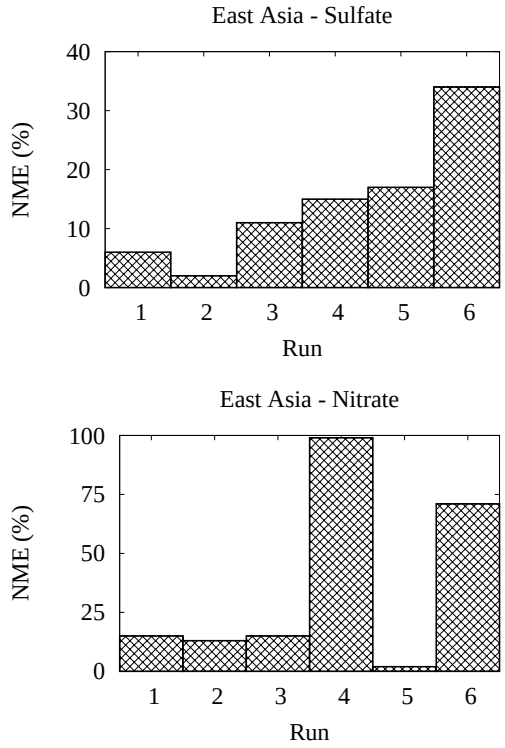

FIG. 3 – Over East Asia, for sulfate and nitrate, differences between the reference run and a run with 3 sections instead of 10 (1), with the hybrid scheme (2), without coagulation (3), with heterogeneous reaction (4), with cloud chemistry (5), using CMAQ instead of Polyphemus-SIREAM (6)

TAB. 4 – Scores for PM_{10}, sulfate and nitrate over Europe 2001. E refers to the EMEP database, A to the airbase database and B to the bdqa database.

	Meas.	Sim.	Corr	NMB	NME
PM_{10} (E)	16.9	15.6	55	-1	51
PM_{10} (A)	24.9	15.4	44	-33	49
PM_{10} (B)	19.8	15.8	57	-19	36
sulfate (E)	2.5	2.1	56	-5	51
sulfate (A)	1.9	2.4	51	73	105
nitrate (E)	2.6	4.1	41	91	122
nitrate (A)	3.5	4.4	72	27	56

tions, without the heterogeneous reaction for N_2O_5, with varying reaction probability for the heterogeneous reaction N_2O_5 (following Evans and Jacob [2005] and Riemer et al. [2003]), with nucleation (by adding 2 bins between 0.001μm and 0.01μm), with the criterion LWC (liquid water content) < 0.07g m^{-3} instead of LWC < 0.05g m^{-3} to call the aqueous model VSRM, with a different criterion to call the aqueous model VSRM (VSRM is called when the cloud fraction is greater than 20%), with different parameterizations of the diameter and falling velocity of raindrops (instead of the parameterizations of Loosmore and Cederwall [2004], the parameterization of Seinfeld [1985] is used for the falling velocity of raindrops and the one of Pruppacher and Klett [1998] is used for the diameter of raindrops), with monthly boundary conditions from Mozart version 2.4 (Horowitz [2003]) instead of daily boundary conditions from Gocart (Chin et al. [2000])), with the parameterization of Louis (Louis [1979]) rather than Troen Mahrt (Troen and Mahrt [1986]) for vertical diffusion.

As shown in Figure 4 for the winter run and Figure 5 for the summer run, PM_{10}, sulfate and nitrate are strongly impacted by the parameterization used for vertical diffusion (for PM_{10}, it corresponds to the highest sensitivity with a NME of 20% in winter and 29% in summer). For PM_{10}, sulfate and nitrate, low sensitivity is observed to the number of sections used, to whether the reaction probability of the heterogeneous reaction N_2O_5 varies or not, to nucleation and the threshold used for LWC. In winter, PM_{10} is sensitive to whether LWC or a cloud criterion is used to call VSRM, to the parameterization of scavenging, to boundary conditions and to the heterogeneous reaction of N_2O_5 (NMEs of about 10%). In summer, PM_{10} is sensitive to boundary conditions (NME = 16%) and to the parameterization used for scavenging (NME = 8%). For sulfate, the highest sensitivity in both winter and summer corresponds to boudary condition (with a NME of 22 and 19%). Sulfate is also sensitive to the criterion used to call VSRM (with a NME of 10 and 8%), and to the heterogeneous reaction N_2O_5 in winter (NME = 8%). For nitrate, there is a high sensitivity to the heterogeneous reaction N_2O_5 (with a NME of 63 and 24%) and to the dynamic mass transfer (with a NME of 17 and 27%). To a lesser extent, nitrate is also sensitive to the cloud criterion use to call VSRM (with a NME of 11 and 16%).

3 CONCLUSION

The chemistry transport model Polyphemus with the aerosol module MAM or SIREAM performs reasonnably well at the regional scale over Paris, Tokyo and at the continental scale over Europe and Asia. For each validation, sensitivity studies were conducted. As expected, PM_{10} is sensitive to configuration

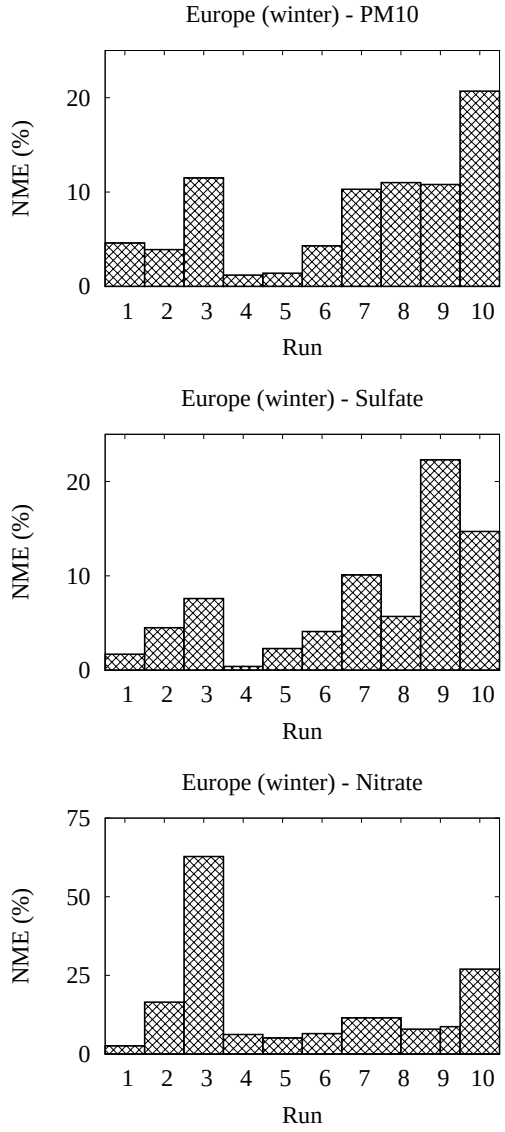

FIG. 4 – Over Europe, for PM_{10}, sulfate and nitrate, differences between the reference run and a run with 10 sections (1), with dynamic mass transfer (2), without the heterogeneous reaction N_2O_5 (3), with varying reaction probability for the heterogeneous reaction N_2O_5 (4), with nucleation (5), with the criterion LWC $< 0.07\mu$g m^{-3} to call the aqueous model VSRM (6), with the cloud fraction to diagnose when to call VSRM (7), with a different parameterization for scavenging (8), with different boundary conditions (9), with a different parameterization for vertical diffusion (10).

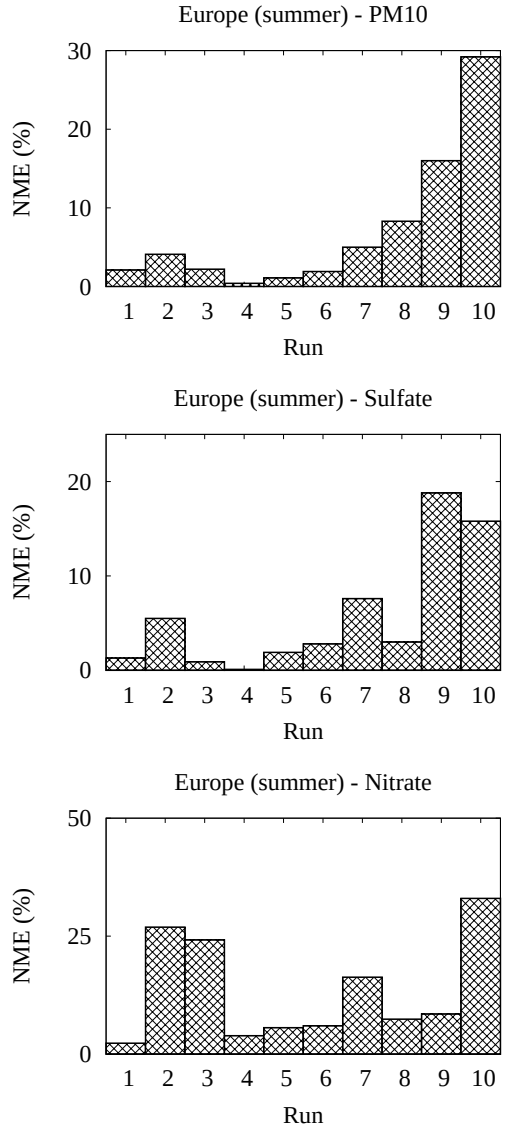

FIG. 5 – Over Europe, for PM_{10}, sulfate and nitrate, differences between the reference run and the other runs of the sensitivity study (see caption of Figure 4).

choices outside the aerosol module, such as vertical diffusion and boundary conditions, but also to parameterizations inside the aerosol module, such as scavenging or heterogeneous reactions. Size distribution, coagulation and nucleation have lower sensitivities. Sulfate is sensitive to boundary conditions, to the criterion used to diagnose clouds. For nitrate, high sensitivity to the heterogeneous reaction of N_2O_5 was observed, although the sensitivity becomes low in the european runs when the reaction probability varies with the aerosol composition, temperature and relative humidity following Evans and Jacob [2005] and Riemer et al. [2003]. The sensitivity to the dynamic mass transfer varies depending on conditions.

RÉFÉRENCES

Chin, M., Rood, R., Lin, S.-J., Muller, J., and Thomspon, A. (2000). Atmospheric sulfur cycle in the global model GOCART : Model description and global properties. *Journal of Geophysical Research*, 105 :24671–24687, 10.1029/2000JD900384.

Debry, E., Fahey, K., Sartelet, K., Sportisse, B., and Tombette, M. (2007). Technical note : A new SIze REsolved Aerosol Model : SIREAM. *Atmos. Chem. Phys.*, 7 :1537–1547.

Evans, M. and Jacob, D. (2005). Impact of new laboratory studies of N2O5 hydrolysis on global model budgets of tropospheric nitrogen oxides, ozone, and oh. *Geophysical Research Letters*, 32 :L09813, doi :10.1029/2005GL022469.

Fahey, K. and Pandis, S. (2003). Size-resolved aqueous-phase chemistry in a three-dimensional chemical transport model. *Journal of Geophysical Research*, 108 :4690.

Horowitz, L. a. (2003). A global simulation of tropospheric ozone and related tracers : description and evaluation of mozart, version 2. *Journal of Geophysical Research*, 108(D24).

Jacob, D. (2000). Heterogeneous chemistry and tropospheric ozone. *Atmospheric Environment*, 34 :2131–2159.

Loosmore, G. and Cederwall, R. (2004). Precipitation scavenging of atmospheric aerosols for emergency response applications : testing an updated model with new real-time data. *Atmospheric Environment*, 38 :993–1003.

Louis, J. (1979). A parametric model of vertical eddy fluxes in the atmosphere. *Boundary Layer Met.*, 17 :187 :202.

Mallet, V., Quelo, D., Sportisse, B., Ahmed de Biasi, M., Debry, E., Korsakissok, I., Wu, L., Roustan, Y., Sartelet, K., Tombette, M., and Foudhil, H. (2007). Technical Note : the air quality modeling system Polyphemus. *Atmos. Chem. Phys. Discuss.*, 7 :6459–6486.

Napari, I., Noppel, M., Vehkamaki, H., and Kulmala, M. (2002). Parametrization of ternary nucleation rates for h_2so_4-nh_3-h_2o vapors. *Journal of Geophysical Research*, 107 (D19) :doi :10.1029/2002JD002132.

Nenes, A., Pandis, S., and Pilinis, C. (1998). Isorropia : A new thermodynamic equilibrium model for multicomponent inorganic aerosols. *Aquatic geochemistry*, 4 :123–152.

Pruppacher, H. and Klett, J. (1998). *Microphysics of Clouds and Precipitation*. Kluwer Academic Publishers.

Riemer, N., Vogel, H., Vogel, B., Schell, B., Ackermann, I., Kessler, C., and Hass, H. (2003). Impact of heterogeneous hydrolysis of n2o5 on chemistry and nitrate aerosol formation in the lower troposphere under photosmog conditions. *Journal of Geophysical Research*, 108(D4, 4144) :doi :10.1029/2002JD002436.

Sartelet, K., Debry, E., Fahey, K., Roustan, Y., Tombette, M., and Sportisse, B. (2007a). Simulation of aerosols and gas-phase species over Europe with the POLYPHEMUS system. part I : model-to-data comparison for 2001. *Atmospheric Environment*. doi :10.1016/j.atmosenv.2007.04.024, in press.

Sartelet, K., Hayami, H., Albriet, B., and Sportisse, B. (2006). Development and preliminary validation of a modal aerosol model for tropospheric chemistry : Mam. *Aerosol Science and Technology*, 40(2) :118–127. doi : 10.1080/02786820500485948.

Sartelet, K., Hayami, H., and Sportisse, B. (2007b). Dominant aerosol processes during high-pollution episodes over Greater Tokyo. *Journal of Geophysical Research*, 112(D14214) :doi :10.1029/2006JD007885.

Sartelet, K., Hayami, H., and Sportisse, B. (2007c). MICS-Asia Phase II : sensitivity to the aerosol module. *Atmospheric Environment*. doi :10.1016/j.atmosenv.2007.03.005, In press.

Schell, B., Ackermann, I., Hass, H., Binkowski, F., and Ebel, A. (2001). Modeling the formation of secondary organic aerosol within a comprehensive air quality model system. *Journal of Geophysical Research*, 106(D22) :28275–28293.

Seinfeld, J. (1985). *Atmospheric physics and chemistry of Air Pollution*. Wiley.

Tombette, M. and Sportisse, B. (2007). Aerosol modeling at regional scale : Model-to-data comparison and sensitivity analysis over Greater Paris. *Atmospheric Environment*. doi :10.1016/j.atmosenv.2006.10.037.

Troen, I. and Mahrt, L. (1986). A simple model of the atmospheric boundary layer : sensitivity to surface evaporation. *Boundary-Layer Meteorology*, 37 :129–148.