

Review of Manuscript “Operational, regional-scale, chemical weather forecasting models in Europe” by J. Kukkonen et al.

General Comments:

This is a very comprehensive review paper on numerical air quality forecasting systems being used in Europe. The authors not only catalog the models but also elaborate on the science process details in each model. This review paper, including its complete citation list, should be a valuable desk reference to all in the operational and research air quality modeling communities for years to come; not only in Europe, but in the global communities where these models are utilized. I have a few minor general criticisms of the manuscript:

1. The subject of the paper is operational air quality forecasting systems, yet neither the text nor the tables list and compare the operational characteristics of the systems. For example, another Table may be needed in which the models are listed along with the institute(s) that use them, the frequency with which forecast simulations are made and public forecasts are issued, the length and temporal resolution of the forecast simulations, the frequency of updates, the spatial domains and grid resolutions used for the forecasts, the methods of forecast product dissemination, and the websites where forecast products are available. Table 12 provides some generic information, but more specifics are needed for each operational model system.

2. The completeness of the model descriptions and of the science process descriptions can be quite variable across models and processes. For example, considerable discussion is devoted to advection/diffusion, and deposition, but relatively little to aerosol physics and dynamics. Perhaps a bit more even treatment is needed.

3. At the risk of making a long manuscript even longer, I think a few more figures are needed to make the manuscript more appealing to the reader. Perhaps some figures illustrating the operational forecast products generated from some of these modeling systems, and several plots illustrating aspects of the science processes would be helpful.

4. Include all acronyms used in the paper in the list of acronym definitions, and refer to the list early in the text. Then any acronym definitions can be removed from the body of the manuscript.

5. Finally, a thorough editorial review is needed. Given the length of the manuscript, there were relatively few errors. Generally the paper is well written; however there are some sections where there are more grammar issues than others.

Specific Comments:

p. 6006 – Provide a reference for the TRAMPER boundary layer module

p. 6011; L25 – It is debatable whether convective parameterizations may be omitted from models using resolutions higher than 5 km. While such was the thinking in the past, many model applications have shown spurious effects of explicit convection with grid sizes of 3-5 km, with

individual convective cells covering too large an area. Some modelers have used convective parameterizations in this range to mitigate the impact of such spurious effects.

p. 6014; L25 – What is the height of the model top of the IFS model?

p. 6018; L7 – Eliminate the phrase “pronounced word, like the Star Trek character”. This reviewer found the reference amusing, but some readers may be mystified by the reference to popular culture.

p. 6019; L8 – change “mixing” to “dilution”

p. 6019; L7-9 – Regarding the assumption of no exchange at the top boundary of the model domain ... This depends on the model vertical structure and the height of the top of the model domain. The statement may not be true for domains of limited vertical extent.

p. 6019; L20-23 – The sequence of processing in the fractional stepping may make a difference in the model results. In other words, whether chemical processing is performed before or after advection/diffusion will give different results. Also, the choice of process where the emissions are injected into the modeling can be a significant choice as this is strong forcing to the model.

p. 6020; L18 – Need to mention here the desirable attributes of being monotonic, positive-definite, and mass-conservative for advection schemes used in CTMs. These attributes are discussed later on pp. 6021-6022, but the discussion of the desirable attributes should be moved forward to p. 6020 before the details of specific methods are described.

p. 6022; L20-23 – It is quite surprising to read that the efficiencies of the advection schemes are less significant than some of the other positive attributes. This may be understandable in research-grade models, but the subject of this paper is Operational models, where efficiency would seem to be at least as important as the other factors.

p. 6023; L5 – Add the CMAQ model to the list of those that use the PPM advection scheme.

p. 6023; L13 – The Plume-in-Grid option is no longer available in the CMAQ model.

p. 6023; L24 – Horizontal diffusion in CMAQ is based on a grid-size-dependent algorithm and uses a combination of Smagorinsky’s approach with a term to minimize numerical diffusion (Byun and Schere, 2006).

p. 6024; L22 – Add the following references for ACM2:

Pleim, J. E., 2007: A combined local and nonlocal closure model for the atmospheric boundary layer. Part I: Model description and testing. *J. Appl. Meteor. Climatol.*, **46**, 1383–1395.

Pleim, J. E., 2007: A combined local and nonlocal closure model for the atmospheric boundary layer. Part II: Application and evaluation in a mesoscale meteorological model. *J. Appl. Meteor. Climatol.*, **46**, 1396–1409.

p. 6026; L28 – “ ... especially if the model domain covers regions with dense forests or agricultural lands.”

p. 6026; L8-9 – Some models have moved from the CBM-IV chemical scheme to the newer CBM05. References:

Yarwood, G., Rao, S., Yocke, M., Whitten, G.Z., 2005. Updates to the Carbon Bond chemical mechanism: CB05. Final Report to the US EPA, RT-0400675, December 8, 2005 /http://www.camx.com/publ/pdfs/CB05_Final_Report_120805.pdf.

Luecken, D.S., S. Phillips, G. Sarwar, C. Jang, 2008: Effects of using the CB05 vs. SAPRC99 vs. CB4 chemical mechanism on model predictions: Ozone and gas-phase photochemical precursor concentrations. *Atmos. Environ.*, 42:5805-5820.

Sarwar, G., D. Luecken, G. Yarwood, G. Whitten, W. Carter, 2008: Impact of an updated Carbon Bond mechanism on predictions from the CMAQ modeling system: Preliminary assessment. *J. Appl. Meteor. Climat.*, 47:3-14.

p. 6026; L23-25; p. 6027 – ISORROPIA really does not belong in this section with other gas-phase chemical mechanisms. It is an inorganic partitioning scheme for aerosols based on thermodynamic principles. You might consider adding a process in the Aerosols section on Aerosol Thermodynamics, and including ISORROPIA there.

p. 6027; L8 – Modify the end of this paragraph: “CBM-IV is widely used in research and regulatory air quality models. Recently an updated version of the Carbon Bond mechanism (CBM05) has become available. Among other changes, this version of the mechanism contains updated rate constants, an extended inorganic reaction set for urban to remote tropospheric conditions, and NO_x recycling reactions to represent the fate of NO_x over multiple days. The CBM05 mechanism has been implemented in the most current versions of the CMAQ model (Sarwar et al., 2008; Luecken et al., 2008).

p. 6032 – Before Section 4.3.9 you should include a section on numerical solvers for gas-phase mechanisms. Just as you discussed the desirable attributes and efficiencies of numerical solvers for advection in an earlier section, there needs to be a similar discussion on chemistry solvers. How the coupled set of chemical differential equations are solved can have significant implications on the accuracy and efficiency of the solution and on the overall model efficiency and results.

p. 6035; L1 – Change “accurate” to “resolved”. Sectional modules for aerosols can provide more resolution to the size distribution, but they are not necessarily more accurate than modal aerosol models.

p. 6035 – The sections on aerosol dynamics are quite thin, especially compared to the sections on gas-phase chemistry. Few details are provided; more references are needed. Here are a few references that could be added:

(Modal scheme)

Binkowski, F.S. and S.J. Roselle, 2003: Models-3 Community Multiscale Air Quality (CMAQ) Model Aerosol Component. I. Model description. *J. Geophys. Res.*, 108, 4183, doi:10.1029/2001JD001409.

(Sectional scheme)

Zhang, K.M., A.S. Wexler, 2008: Modeling urban and regional aerosols-Development of the UCD Aerosol Module and implementation in CMAQ model. *Atmos. Environ.*, 42:3166-3178.
Zhang, Y., B. Pun, K. Vijayaraghavan, S.-Y. Wu, C. Seigneur, S.N. Pandis, M.Z. Jacobson, A. Nenes, J.H. Seinfeld, 2004. Development and application of the model of aerosol dynamics, reaction, ionization and dissolution (MADRID). *J. Geophys. Res.*, 109, D01202, doi:10.1029/2003JD003501.

p. 6040 – Somewhere in the section on dry deposition, it should be mentioned that it is quite difficult to evaluate this process in the atmosphere due to a lack of direct measurements of deposition fluxes.

p. 6043; L3-4 – This section should also acknowledge the difficulties in generating accurate precipitation amounts and location (especially from convective systems) from the meteorological models. The biases in the precipitation predictions will greatly affect the wet deposition estimates.

p. 6043; L15-16 – While you do acknowledge that anthropogenic emissions are not addressed in detail here, it gives the impression that a major aspect of the forcing of the CWF models is missing from this review. Consider adding a few sentences acknowledging the primary forcing aspects of such emissions on CWF models before you indicate that they are beyond the scope of this review. (Perhaps provide a few references to the major European inventory descriptions?)

p. 6044; L24-26 – In addition to the factors mentioned, the various tree and crop species and their respective emission factors also are key to natural emissions estimation.

p. 6054; 21 – Add the following reference to the list of model evaluation overviews:
(Dennis et al., 2010):

Dennis, R., T. Fox, M. Fuentes, A. Gilliland, S. Hanna, C. Hogrefe, J. Irwin, S.T. Rao, R. Scheffe, K. Schere, D. Steyn, A. Venkatram, 2010: A framework for evaluating regional-scale photochemical modeling systems. *Environ. Fluid. Mech.*, 10:471-489.

p. 6056; L13-15 – This depends upon the density and adequacy of the observational networks.

p. 6057; L11 – Change “validation” to “evaluation”. These terms are not synonymous.

p. 6057; L19 – Modify the following sentence: “... 2003), the EU ENSEMBLE project ..., and the on-going Air Quality Model Evaluation International Initiative (AQMEII; Rao et al., 2011).”
Reference: Rao, S.T., S. Galmarini, K. Puckett, 2011: Air Quality Model Evaluation International Initiative (AQMEII) – Advancing the state of the science in regional photochemical modeling and its applications. *Bulletin of the American Meteorological Society*, 92:23-30.

p. 6061; L8 – Add CMAQ to the list of open-source models.

p. 6061-6062 – Somewhere in Section 6 should be discussed the important role of user support and training for the publicly available models.

p. 6076; L5-7 – Informing emission inventory corrections via (passive; retrospective) data assimilation may go even further than dynamic chemical data assimilation during the model initialization period, since the emissions forcing proceeds throughout the entire model simulation, not just during initialization.

p. 6079; L1-13 – While it is important to note these possible improvements in the cloud microphysics schemes, one should also note that it is even more important to estimate the clouds in the right place at the right time in the meteorological model. Otherwise the added complexity of the cloud description is wasted.

p. 6079-6080 – Missing processes: deposition, gas-phase species (including air toxics chemistry). Do you want to mention any needed improvements to these processes?

p. 6080-6081 – The most important reason for model evaluation is to build confidence in the model's use for a particular application. This should be stated early in this section.

p. 6082; L16 – Change “validation” to “evaluation”. Validation implies the soundness of the underlying basis of the model in transforming the basic science into the model's equations. Evaluation is the process of comparing the model results with measurements.

p. 6082 – Suggest acknowledging the role of intensive data collected in special field campaigns to assist diagnostic evaluation of model processes

p. 6086; L10-13 – Add to this list the lack of sufficient chemical/aerosol measurements in the vertical dimension.

p. 6086; L24-26 – Such multi-model ensembles might be called “ensembles of opportunity”, as distinguished from classical ensembles where the same model is used with different initial conditions.