

25 July 2011

Review of *Atmospheric Chemistry and Physics Discussions* manuscript no. acp-2011-40

entitled

***“Operational, regional-scale, chemical weather forecasting models in Europe”***

by

J. Kukkonen, T. Balk, D.M. Schultz, et al.

## GENERAL COMMENTS

The authors' goal for this paper was to provide a systematic assessment and comprehensive review of the current state of the science of operational, regional-scale chemical weather forecasting in Europe, and they have largely succeeded in their attempt. The resulting document is encyclopedic: 105 pages of text, 47 pages of references, and 26 pages of figures and tables, for a total of 178 pages.

This is a generally well-written and useful paper and it provides a good overview and valuable synthesis of the subject. The organization of the paper is logical and comprehensive. The text includes useful comparisons and contrasts with numerical weather prediction, and the value of the paper is enhanced by a concluding discussion of emerging research and development areas and future challenges for chemical weather forecasting in Europe and elsewhere. A notable strength of the paper are the detailed model comparison tables that are provided. While the paper is much longer than usual, I think the length can be justified given the subject, the goal of the paper, and the large amount of information that has been synthesized in one place together based on expert judgement and perspective.

Nonetheless, in reviewing this paper I encountered a number of “rough spots” where the discussion was unclear or seemed to be inconsistent or incomplete or uneven. I have made a number of specific comments and suggestions below that I would ask the authors to consider, as I believe that addressing them would strengthen the paper further by improving its clarity, consistency, completeness, and balance. I have also included many editorial comments to improve the text, but their number is perhaps to be expected with a manuscript of this length.

## SPECIFIC COMMENTS

1. This is an unusually long manuscript, but the current title does not suggest this length or the reason for it. On page 5989, lines 4-5, the authors state that the aim of the article is “to bring the field up to date with the most comprehensive summary and assessment of the state of CWF”. Accordingly, I would suggest that the current title be expanded to indicate this aim by adding an introductory phrase such as “A review of ...”, “A survey of”, or “A summary and assessment of”.

2. In such a long manuscript, clarity and consistency of terminology are very important. However, I found there to be some imprecision and inconsistency in some basic and important terminology in the context of this paper:

- the term “chemical weather forecasting models” is used in this paper [e.g., p. 5987, l. 3], but the terms “atmospheric chemistry models” [e.g., p. 5987, l. 6], “chemical transport models” [e.g., p. 5987, l. 17] “air-quality forecasting models” [e.g., p. 5988, l. 2], “regional air-quality models” [e.g., p. 5990, l. 17], “dispersion models” [p. 5992, l. 25 and Table 1], “air-chemistry models” [e.g., p. 6018, l. 19], and “ACT models” [p. 6067, l. 13] are also used, apparently interchangeably.

I would suggest that the authors either state early on that all of these terms are synonymous or else restrict their usage to just a few of them.

- the terms “models”, “systems”, and “modelling systems” [e.g., p. 5988, p. 5994] are also used throughout the paper, again apparently interchangeably.

For example, consider the first sentence of the Abstract vs. the title of Section 2.2:

“Numerical models that combine weather forecasting and atmospheric chemistry are here referred to as chemical weather forecasting models” – *suggests that CWF “models” are modelling systems that consist of NWP and CTM components in either an off-line or on-line framework*

“The integration and coupling of NWP and CWF models” – *suggests that a CWF model is a CTM*

Moreover, it is suggested by Table 1 that 15 of the 18 European CWF modelling systems considered in the manuscript are off-line modelling systems and only three are on-line models. One solution would be to follow Section 2.2 and refer in the manuscript to CWF *modelling systems* that are composed of NWP models and CTMs coupled in either an off-line or on-line framework. This would require a change to the title, to the first line of the abstract, and so on, and the title of Section 2.2 would be changed to “The integration and coupling of NWP and chemical transport models”.

I would also suggest avoiding the term “dispersion model” if chemistry is a represented process (see p. 5992, l. 25 and Table 1).

3. Re p. 5988, l. 28-29 and Section 5.3 – the Rao et al. (2011) *Bull. Amer. Meteor. Soc.* article on the AQMEII model intercomparison study could be referenced as a example of such a scientific evaluation that will soon be available to the community.
4. A key aspect of the CWF modelling systems considered in this manuscript is that they are “operational”, but I was not able to find an explicit definition of this term in the manuscript. Perhaps the closest the authors come is at the top of page 5992, where operational CWF modelling systems are defined by exclusion, i.e., by what they are not. Table 12. on the other hand, lists some properties of these systems, but this is not the same as a definition. Presumably the characteristics of an operational CWF system include the routine preparation of short-term forecasts that made every day, disseminated automatically to some clients, and take less time to prepare and issue than real time so that the forecasts are still useful (i.e., still timely, still referring to the future) when they are received. I also noticed on p. 5990 that the

AQ modelling systems involved in the GEMS project are “quasi-operational”, but no explanation of the differences between “operational”, and “quasi-operational” is given.

5. Other European operational AQ forecast models that have not been selected for this summary such as CALIOPE and GEM-AQ (<http://ecoforecast.eu/>) could be mentioned in Section 2.1.
6. Section 2.2 notes (p. 5995, l. 2) that there are “potential problems communicating between off-line coupled meteorological and CWF models”. Presumably these include the very serious potential problems with mass consistency and mass conservation discussed by Byun (1999a,b), which would be well worth referencing. It is also stated in this section that only two of the 18 CWF modelling systems are on-line systems, but Table 1 indicates that SKIRON/Dust is also an on-line system. Is the difference the implementation (or not) of two-way interactions?
7. The authors note on page 5988 that syntheses of information about CWFISs are scarce and note on page 5989 that a systematic review of CWF modelling systems would be very valuable in assessing the strengths and weaknesses of these systems. I believe the goal of this manuscript is to provide such a systematic review of European CWF modelling systems, but the CWF modelling system descriptions presented in Section 2.3 are anything but systematic and do not provide a consistent minimum set of relevant characteristics and properties across all of the model descriptions. Such a minimum set of characteristics should include specific operational characteristics and could include information on the following characteristics for all 18 CWFIS modelling systems:
  - meaning of CWFIS acronym
  - names of CWFIS component NWP model and CTM
  - modelling system developer
  - agency running CWFIS operationally
  - coupling method
  - source of anthropogenic and biogenic emissions
  - treatment of chemical lateral boundary conditions
  - horizontal grid spacing, vertical grid spacing, and height of model top
  - forecast species
  - forecast frequency
  - forecast duration
  - method(s) of forecast dissemination

It might be argued that this information is provided in the accompanying summary tables, but that is only true for some of these characteristics. For example, none of the tables provides a summary of operational characteristics for the 18 modelling systems even though operational models are the focus of the manuscript. It might be also be argued that including such a minimum set of characteristics would lengthen the descriptions further, but the shortest modelling system description (Section 2.3.8) actually provides most of the above information, suggesting that the amount of additional text would be modest.

8. There are some apparent discrepancies between Section 2.3, Section 4, and the summary tables:

- Section 2.3.4 discusses both on-line and off-line versions of Enviro-HIRLAM but Table 1 only mentions the on-line version;
  - Section 2.3.3 mentions that EURAD-RIU dry deposition “depends upon the particle itself” [p. 5998, l. 5] but the EURAD-RIU particle dry deposition scheme is not described in Table 6; also, EURAD-RIU is not listed in Table 8a for gaseous natural emissions; even if it does not consider any natural emissions, that fact should be noted in the table.
  - Section 2.3.11 mentions that MOCAGE predicts aerosols [p. 6004, l. 11] and a short description of the MOCAGE aerosol package is included in Table 3, but no information about the MOCAGE aerosol scheme is included in Table 6.
9. Anthropogenic emissions are crucial for CWF modelling systems (as noted on p. 6018, l. 24-25), but only some of the modelling system descriptions in Section 2.3 mention this topic and no summary table is provided for model treatment of anthropogenic emissions even though summary tables are provided for natural emissions (Tables 8a, 8b). This neglect seems to have been deliberate, but the text does not mention any decision not to discuss anthropogenic emissions until Section 4.6 and even then provides no justification for this decision. Other than describing which emissions inventories have been used, other process-related representational aspects of the treatment of anthropogenic emissions that are also not discussed are whether each CTM differentiates between ground-level and elevated emissions, whether plume rise is modelled, whether subgrid-scale plumes are modelled, what tools have been used to prepare emissions for CTM use, and what techniques have been employed for spatial disaggregation, temporal disaggregation, PM size disaggregation, and chemical speciation.
  10. I would suggest that the discussion in Section 3.1 be cast in terms of “*selected* physical processes” (cf. Table 2 title). For example, other physical processes considered by NWP models such as shortwave and longwave radiation, surface-layer physics, land-surface processes, and gravity wave drag are not discussed, and Section 3.1.5 neglects the topic of upper boundary conditions. It would be useful to mention as well why this particular set of NWP processes was selected for a review paper on CWF modelling systems.
  11. Section 3.1.3 notes that convective parameterization schemes redistribute heat and moisture. However, a number of global chemical transport models employ modified convective parameterization schemes to treat the subgrid-scale vertical transport of chemical tracers by deep convection. Do any of the modelling systems reviewed in Section 2.3 consider this process?
  12. Concerning lateral boundary conditions in NWP models (Section 3.1.5), Staniforth (1997, *Meteorol. Atmos. Phys.*, 63, 15-29) provided both a theoretical and practical review of this topic and proposed an “acid test” for any LBC scheme.
  13. Are Section 3.3 and Table 2 missing some relevant meteorological models? The discussion of FARM (Section 2.3.5) mentions the RAMS meteorological model and the discussion of SILAM (Section 2.3.15) mentions the AROME meteorological model.
  14. Re “It is typical to assume that ... there is no exchange at the top boundary of the domain” [p. 6019, l. 12-14]: depending on the location of the model top, this assumption might preclude representation of stratospheric intrusions of ozone.

15. Section 4.2 lacks the introductory discussion provided for other topics in Section 4. Some possible topics could include (a) difficulties in parameterizing vertical diffusion in stably-stratified boundary layers, (b) the appropriateness of including an explicit treatment of horizontal diffusivity given the well-known numerical diffusion associated with many advection schemes, (c) the limited treatment of free-tropospheric vertical diffusion typically found in CWFIS models, (d) schemes that consider enhanced diffusivity at the top of cloud-topped boundary layers, and (e) subgrid-scale tracer transport in deep convection (given that, fundamentally, vertical diffusion may be viewed as convection by unresolved subgrid-scale flows, including flows in cloud systems).
16. In Section 4.3, it would be helpful to the reader if the implication of the phrase “depending on the focus of the modeling system” [p. 6026, l. 2] were explained. Presumably what is meant is that (a) a representation of aqueous-phase sulphur chemistry is required if a CTM is to be used for acid deposition and acidification/eutrophication studies and (b) a representation of inorganic gas-particle partitioning is required if a CTM is to be used to predict atmospheric PM.
17. How does the UNI-OZONE mechanism treat inorganic thermodynamic equilibrium (Section 4.3.8) given that neither the EMEP model or MATCH use the ISORROPIA scheme (cf. Section 4.3.2)?
18. Despite this manuscript’s 47 pages of references, no references are given in Sections 4.4.1, 4.4.2, 4.4.3, or 4.4.4, but the inclusion of some references in these sections would be very useful for the reader. For example, where can the reader find out more about the modal approach or about EQSAM? Section 4.4 should also include some mention about major PM chemical components (with a reference to the 10 components listed in Table 6) and model assumptions about particle mixing state (internal vs. external). Section 4.4.2 could note the need for periodic mode redistributions. The comment in Section 4.4.3 regarding the challenge of providing size-distributed PM emissions also applies equally to the chemical speciation of PM emissions. And it could be noted in Section 4.4.4 that some microphysical processes depend on particle number, some on particle area, and some on particle volume.
19. In Section 4.5.1, in the paragraph on p. 6038 about the parameterizations of  $R_a$  and  $R_b$ , if 10 of the 18 models follow one approach, what can be said about the other eight models? And regarding  $R_c$ , Zhang et al. (2002, *Atmos. Environ.*, **36**, 537-560) among others have discussed the influence of the physico-chemical properties of depositing gaseous species on this resistance term.
20. Concerning the influence of rainfall and particle properties on particle scavenging discussed on p. 6042 of Section 4.5.2, Wang et al. (2010, *Atmos. Chem. Phys.* **10**, 5685-5705) recently provided a systematic examination of all of the factors related to rain and aerosol particle properties on below-cloud scavenging of size-resolved particles by rain. This paper may at least partly address the issue raised in the last sentence of the section.
21. Are the lightning NO<sub>x</sub> emissions used by THOR (Section 4.6.1) and the wildfire emissions used by THOR (Section 4.6.2) for operational forecasting climatological in nature, and if so, why is the use of such emissions better than neglecting them entirely if it is not possible to assign the emissions to a specific time and place and magnitude?

22. For Aeolian dust emissions or fugitive dust emissions from agriculture, construction, and other activities, is any consideration given in any of these European AQ models (or emissions processing models) to subgrid-scale near-source removal by sedimentation or impaction (cf. [http://www.epa.gov/ttn/chief/emch/dustfractions/transportable\\_fraction\\_080305\\_rev.pdf](http://www.epa.gov/ttn/chief/emch/dustfractions/transportable_fraction_080305_rev.pdf))?
23. I am grateful to the authors for their discussion in Section 4.7 of the difference between grid spacing and resolution. This is a distinction that is too often confused or misunderstood. My only comment is that the linkage between Eulerian grid spacing and Lagrangian models is also not widely understood. It might be worth adding a few lines to this section to note that the effective grid spacing of the NAME model is determined by the grid spacing of the emissions and especially the meteorological fields that NAME uses. The more general conclusion for both Lagrangian and Eulerian models is that the “effective resolution” of the modelling *system* is probably determined by the coarsest of the three grids that are employed by the NWP model, the emissions processing model, and the CTM.
24. There is no discussion in Section 4 of the topic of chemical initial conditions and lateral boundary conditions comparable to the discussion for NWP models given in Section 3.1.5. The lack of discussion on chemical lateral boundary conditions is an important gap given that all 18 of the CWF models described in this manuscript are limited-area models.
25. The authors would provide a service to their readers if in Section 5 they defined and distinguished between such terms as evaluation, verification, and validation (cf. Fox, 1981, *Bull. Amer. Meteor. Soc.*, **62**, p. 600). As George Box famously wrote, “All models are wrong. Some models are useful.” Testing the *validity* of a model (p. 6051, l. 25) is clearly nonsensical if we accept that a model can only be an imperfect representation of the real world. Rather than “to test their validity”, I would suggest that the objective is “to evaluate their skill and usefulness”. It is worth noting that for historical reasons the NWP community uses a different terminology, including “verification” in place of “evaluation”. To avoid confusion, it would be helpful to replace the use of “verification” on page 6056 with “evaluation”.
26. I believe Section 5.1 would benefit from several revisions.
- Given that this section of necessity provides only a very condensed overview, the reader would benefit from the inclusion of some judiciously chosen references to provide sources of additional information: there are numerous possibilities, including Dunker et al. (2002a,b, *ES&T*), Hakami et al. (2003, 2006, *ES&T*), Koo et al. (2007, 2009, *ES&T*), Napelenok et al. (2006, *Atmos. Environ.*), Zhang et al. (2005, *JGR*, D02305), ...
  - To help distinguish sensitivity analysis (SA) from evaluation, it could be noted in this section that SA does NOT require the use of observational data.
  - SA is defined in the first paragraph of the section as being the study of model output response to variations in model inputs. But then, confusingly, backward SA and adjoint sensitivities are discussed on page 6053. This inconsistency could be avoided if the word “forward” were substituted at the very beginning of Section 5 or if the first sentence of the section were extended by adding the phrase “and the variation of model inputs resulting from variation of model outputs”.
  - It would help the reader if synonymous terms were mentioned. For example, in the discussion of the statistical and deterministic approaches to SA at the bottom of p. 6052,

relevant or equivalent terms like Monte Carlo analysis, tangent linear model, and direct decoupled method could be mentioned.

- The basic structure of this manuscript is to begin with a general discussion and then follow with a specific discussion of the 18 models being considered. In this section, a paragraph could be appended that discusses published SA studies performed with any of these 18 models.

27. It would be worth noting in the first paragraph of Section 5.2 that evaluations should be application-specific (cf. Dennis et al., 2010, *Environ. Fluid Mech.*, **10**, 471-489) given that this expectation is alluded to in the second paragraph. And in the second paragraph, besides biases and errors, another relevant metric is model skill in reproducing patterns (i.e., correlations).

28. In Section 5.2 two generic evaluation protocols are introduced (general-scientific-benchmark-operational and operational-diagnostic-dynamic-probabilistic) but only the second one is discussed. The different approaches of the first protocol should either be defined in the text or any mention of the first protocol deleted from the text.

29. Section 5.2 discusses the comparison of model predictions with measurements, including different categories of evaluation, but it does not similarly distinguish between different categories of measurements. It would be worth noting that there are two main categories of measurements, that is, (a) ongoing routine network measurements and (b) short-term higher-resource campaign measurements, and that these different types of measurements have different uses, advantages, and limitations.

30. On p. 6056 (l. 18) of Section 5.2, there is an abrupt introduction of “the classification”: What classification? Classification of what?. Presumably it is the classification of “Evaluation Level” used in Table 10, but there is no link made between this paragraph and that table. Furthermore, a website [[http://pandora.meng.auth.gr/mds/long\\_help.php](http://pandora.meng.auth.gr/mds/long_help.php)] is given as the source of this classification, but it turns out that considerable though unmentioned revisions have been made to the following original level definitions:

- This level of evaluation is hard to achieve because of either still pending work on evaluation, or minor limitations in the measurements available (quality, representativeness, coverage etc.), or both.
- Extensive and good model evaluation has been performed, but still uncertainties because of major limitations in the measured data.
- Considerable uncertainties because of both lack of measurements and an inadequate evaluation procedure.
- Only first attempts towards evaluation.
- No evaluation at all.

Moreover, the revisions to the level definitions that are provided on pages 6056 and 6057 are not complete and mutually exclusive. For example, in which level would a probabilistic analysis go? And a comprehensive and well done operational evaluation would seem to fall somewhere between levels 2 and 3 but not belong to either.

Also, how were the entries given under “Evaluation level” in Table 10 assigned? Given the many apparent inconsistencies between models, my suspicion is that they were self-assigned

by each modelling group. The source of these assignments should be mentioned in the text to help the reader judge their accuracy. The many forms of EuroDelta and CityDelta in this table should also be made consistent with Section 5.3, and if the column header “References (up to 4)” is to be believed, the MM5-CAMx group should be asked to delete one reference.

31. There are a few aspects of Section 5.3 that should be reconsidered:

- Mentioning two passive-tracer field experiments, ETEX and Kincaid, the second of which was also a short-range experiment, without any caveats in a review of regional-scale chemical weather forecast models seems misleading. These are useful evaluation data sets for the evaluation of some process representations (i.e., emissions, transport-diffusion) but not others (e.g., chemical transformation, dry deposition).
- Is it appropriate to mention a climate-change ensemble model study (EU ENSEMBLE project) in the context of this review?
- The three multi-model intercomparison studies summarized in this section were selected in part because they included models reviewed in the present manuscript. However, it is not always obvious which models these are. In Section 5.3.1, seven models are listed and one (TM5) is singled out as not being a regional CWF model. But it was not obvious to me that the DEHM (THOR?) and Unified EMEP models (?) are considered in this manuscript (cf. Table 1). On the other hand, in Section 5.3.2, it says that “REM (RCG)” is not included in this article, but RCG is in fact listed in Table 1. The EMEP model is mentioned as well, again with the implication that it is included in this manuscript.
- This may be a naïve question, but since the GEMS-RAQ project (<http://gems.ecmwf.int/do/get/Themes/RAQ>) included the NAME-AQ, CHIMERE, SILAM, MOCAGE, EMEP, EURAD, MATCH, and CAMx models and was focused on operational AQ forecasting, why does this section not include a summary of results and findings from the GEMS-RAQ operational evaluations?
- It could be noted in this section that multi-model evaluation results will soon be available from the AQMEII model intercomparison study.

32. Comment 30 above applies equally in Section 6.1. Table 11 includes a column on “Documentation status” that is expressed in terms of different levels, but these levels are not defined or even mentioned in Section 6.1 and no link is made in the text to Table 11.

33. The scope of Section 6.2 appears to wander in a few places. The introduction to Section 6 indicates that the focus of the section is on different users of *operational* AQ forecasts, but then Section 6.2.1 includes a paragraph discussing emissions scenarios. Section 6.2.2 also mentions “scenario-based estimates” but then goes on to explain that such simulations allow policymakers to design short-term abatement strategies beforehand so as to be ready if the operational forecasts predict unacceptable air quality conditions. This same explanation could be added if appropriate to Section 6.2.1 or else the paragraph on emissions scenarios could be deleted.

34. The discussion in Section 6.2 does not discuss the importance of different measures of model skill to different user communities. For example, for the categorical statistics of hits, misses, and false alarms, it would be interesting if the authors would characterize how different user communities view the relative importance of misses vs. false alarms.



35. The title of Section 6.2.4 might be expanded to “The general public and susceptible populations”, where the text would note that “susceptible populations” includes children, the elderly, and adults with respiratory or cardiovascular impairments.
36. Given the scope of this manuscript, it would be very appropriate for the authors to state at the end of Section 6.3 which forecast products they believe are the most important to disseminate over the Internet for various user communities and how important it is for users to have access to past forecast products from previous days or weeks.
37. For Section 7.1.1, there are a few more points that could be considered.
- The text (p. 6068, line 7) emphasizes particle EC and primary OC, but Table 6 also lists other PM chemical components of interest (sea salt, dust, SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>). All are needed for particle mass closure, for determining a particle’s physical and chemical properties, and for comparison with speciated PM concentration and precipitation-chemistry measurements. An issue specific to primary OC is the need to provide information about organic matter (OM) fraction as opposed to OC fraction for mass closure and receptor analysis. A recent paper by Reff et al. (2009, *ES&T*, **43** (15), 5790-5796) provides a good summary of the state of the science of speciated PM emissions.
  - Recent research suggests that inventories are also missing emissions of numerous semi-volatile and low-reactivity organic compounds (SVOCs and IVOCs) that are neither particle-phase compounds nor VOCs but which contribute to SOA formation.
  - Is it appropriate to discuss *inventories* of biogenic emissions rather than *models* of biogenic emissions given the dynamic nature of biogenic emissions and their strong dependence on meteorological and other environmental conditions?
  - Another issue specific to fugitive dust emissions is the need to treat near-source removal by settling and impaction, which are subgrid-scale processes (e.g., [http://www.epa.gov/ttnchie1/emch/dustfractions/transportable\\_fraction\\_080305\\_rev.pdf](http://www.epa.gov/ttnchie1/emch/dustfractions/transportable_fraction_080305_rev.pdf)).
38. Three additional points might be mentioned in Section 7.1.2:
- Providing pre-gridded emissions inventories is an obstacle for modelling applications where a smaller grid spacing will be used (e.g., nested urban-scale modelling) and introduces the potential for interpolation errors even when these emissions are used with a model grid based on a larger or comparable but different grid spacing, a different grid orientation, and a different map projection;
  - The SNAP source-classification scheme used in many European inventories may be too crude to allow the use of detailed libraries of temporal profiles and speciation profiles for temporal allocation and chemical speciation. In contrast the U.S. EPA’s source classification scheme includes roughly 5,000 values.
  - For operational AQ forecasting, the available inventories are always retrospective and never current, so adjustments are needed (but seldom available) to project the available retrospective inventory forward to the current year of interest
39. The discussion in Section 7.2 on interface modules between NWP models and CTMs is important and worthwhile but it neglects numerical issues such as mass inconsistency and mass conservation problems that can also arise due to interpolation (e.g., Byun, 1999a,b; cf. comment 6).

40. In Table 1, a better choice for the column entitled “Dispersion model” would be “CTM” or “AQ model” or “CWF model”. As per the statement on p. 6050, l. 4, it would be useful to include the height of the model top in the “Vertical grid spacing” column (it is given already for some models). For the NAME model, what is meant by “Continuously variable” in terms of number of levels and top of model (same comment for Table 9)?
41. In Table 3 the advection scheme used is not given for all models (see ALADIN-CAMx, CAMx-AMWFG, MM5-CAMx). It also seems inconsistent that no description is given of the ALADIN-CAMx aerosol package in Table 3 whereas Table 6 has an extensive description. Also, for NAME what does “advection ascheme” mean (would “advection fields” be better)?
42. In Table 9, four of the five columns largely duplicate information given in Table 1. Is Table 9 necessary, or could Table 1 instead be shortened or else other information substituted in Table 1 in place of the columns duplicated in Table 9? Also, the entries under “Type/Coordinate System” are uneven in terms of level of detail, with some mentioning map projections and even grid-cell stencils and some not.

#### TECHNICAL AND TYPOGRAPHICAL CORRECTIONS

- p. 5987, l. 8 “... architecture affects ...”
- p. 5987, l. 13 Many uses of both “modelling” and “modeling”
- p. 5989, l. 5 Suggest “... date with a comprehensive summary and assessment of the state of CWF *in Europe*”
- p. 5989, l. 6 Perhaps “... modelling and forecasting”
- p. 5989, l. 22 Rather than “operational”, perhaps “available”?
- p. 5990, l. 2 Perhaps “There are several other prominent ongoing European projects ...”
- p. 5991, l. 8 “Tørseth and Fahre Vik”? (see p. 6132)
- p. 5991, l. 10 Re “huge variety”, you state on p. 5988 that there may possibly be more than a hundred CWFISs – is 100 really a “huge” number?
- p. 5991, l. 19 “evaluate preliminarily” is awkward – can you reword?
- p. 5992, l. 9 Perhaps “... CWF modelling systems and gives an overview of some of ...”
- p. 5992, l. 25 Do you really mean “adjoint (inverse) *dispersion* modelling” or a term suggesting that chemistry is also considered?
- p. 5993, l. 1 Perhaps “Introduction to Operational CWF Modelling Systems”
- p. 5993, l. 2-7 In the first paragraphs of Sections 5, 6, and 7, a short summary of each subsection is given by subsection number, but this structure is not followed at the beginning of Sections 2, 3, and 4. Can you standardize the structure of these introductory paragraphs?
- p. 5994, l. 6 Both “off-line” and “offline” are used throughout the manuscript.

- p. 5994, l. 12 Both “on-line” and “online” are used throughout the manuscript.
- p. 5995, l. 2 Change “lead” to “led”
- p. 5995, l. 15 Perhaps “Overview of the CWF modelling systems”
- p. 5995-6010 Some of the CWF modelling system descriptions in Section 2.3 (Sections 2.3.1, 2.3.3, 2.3.8, 2.3.14, 2.3.16, 2.3.17) provide links to meteorological model descriptions in Section 3.2 but some (e.g., Sections 2.3.11, 2.3.10 for UK, 2.3.13) do not even mention which meteorological model is employed. This latter gap should be addressed and more links added between Sections 2.3 and 3.2.
- p. 5997, l. 7 To maintain alphabetical order, should EURAD-RIU and Environ-HIRLAM sections be switched (cf. Table 1 ordering)?
- p. 5998, l. 26 “chemical mechanisms” would be preferable.
- p. 6009, l. 10 “Computer Science”?
- p. 6010, l. 5 Either “divided” or “separated” would read better than “broken”.
- p. 6010, l. 20 Perhaps “size distributions and number concentrations”?
- p. 6013, l. 8 “stably stratified situations”?
- p. 6014, l. 15 This title duplicates the Section 3 title; it could be expanded to something like “NWP models used in European CWF modelling systems”
- p. 6014-18 Section 2.3 presents descriptions of the 18 CWF modelling systems in alphabetical order as does Table 1. Table 2 lists meteorological models in alphabetical order. Why not arrange the meteorological model summaries in Section 3.3 in alphabetical order as well?
- p. 6014, l. 23 Is ECMWF model still T1279 as of June 2011?
- p. 6014, l. 25 It would be useful to give the height or location of the model top here as well.
- p. 6015, l. 18 Is the global version hydrostatic or non-hydrostatic? Perhaps “hydrostatic global model or a nonhydrostatic limited-area model”?
- p. 6018, l. 19 There is a terminological disconnect between the section title and the first sentence of the section.
- p. 6019, l. 2 Rather than “interactions”, perhaps “... four types of processes”?
- p. 6019, l. 20 Perhaps “model construction employs operator splitting (e.g., Seinfeld ...)”?
- p. 6020, l. 4 Should be “Richtmeyer” (also p. 6123, l. 14)
- p. 6020, l. 20 Should be “Smolarkiewicz” (also p. 6127, l. 27)
- p. 6020, l. 20 Should be “Rasch” (also p. 6023, l. 7, p. 6134, l. 31, Table 3)
- p. 6020, l. 21 Should be “Côté” (also p. 6021, l. 20 and p. 6128, l. 33)

- p. 6022, l. 17 “Two features ... have somewhat outstanding importance” is awkward – perhaps “Two other features are also important”?
- p. 6022, l. 29 Rather than “solutions”, perhaps “advection scheme”?
- p. 6023, l. 19 “Diffusion is treated fully” – need to give more detail on what is meant by “fully”
- p. 6025, l. 24 Perhaps “... mechanism of VOCs is necessary in any CWF ...”?
- p. 6026, l. 8 Replace RADM2” by “RADM and RACM”? (cf. p. 6029, l. 7-8)?
- p. 6026, l. 28 Should this be “lumped-structure”?
- p. 6029, l. 18 Sentence needs a predicate.
- p. 6030, l. 25 It seems odd that this scheme, which is given its own section, is then not listed on p. 6026, l. 8 or included in Table 4
- p. 6033, l. 6 “Aerosol particles” would be a more accurate title; same comment applies to Table 6 title.
- p. 6033, l. 23 “... include only a fraction of the particulate matter components” might be better.
- p. 6036, l. 7+ The first two sentences of this paragraph seem inconsistent, in that the first sentence states that bulk schemes only consider deposition but the second sentence discusses condensation/evaporation, which bulk schemes also consider.
- p. 6036, l. 22 Perhaps “..., however, can also limit short-term forecasts of ground-level ...”?
- p. 6036, l. 2-5 The first two sentences of this paragraph could be combined since dry deposition is governed by all three of these factors.
- p. 6037, l. 7 Is it necessary to reference both Seinfeld and Pandis (1998) and Seinfeld and Pandis (2006) in different places in this manuscript?
- p. 6037, l. 14 Perhaps “vegetation canopies”?
- p. 6038, l. 2 For clarity, perhaps “... arise from the way the CWF models are interlaced ...”
- p. 6038, l. 20 Instead of “lower canopy resistances”, perhaps “other canopy structures”?
- p. 6041, l. 2-3 “earth’s surface”
- p. 6041, l. 15 “... could potentially be sufficient to ??? CWF models”?
- p. 6041, l. 23 “spatial”
- p. 6043, l. 19 Do the authors really want to say that ozone, PM, and SOA are “anthropogenic air pollutants”, or rather are these air pollutants that have both natural and anthropogenic sources?!
- p. 6044, l. 9 Is Vautard et al. (2005) an appropriate reference for the U.S.? Another relevant study for the U.S. is Park et al. (2010, J. Geophys. Res., **115**, D16210, doi:10.1029/2009JD013144).
- p. 6045, l. 5 “Almost all *of the* 18 CWF models use ...” would be more precise.

- p. 6045, l. 20 Rather than “except for”, don’t you mean “in addition to”?
- p. 6046, l. 6 Perhaps “Other particulates are formed by way of ...”?
- p. 6046, l. 11 Is the second sentence of this paragraph redundant?
- p. 6049, l. 12 Perhaps “Resolution commonly refers to the *smallest* spatial or temporal scale ...”
- p. 6050, l. 22 Perhaps “A summary of the *computational* grid spacings and ...” (consider that the previous paragraph describes the many grid spacings relevant to an AQ modelling system)
- p. 6050, l. 25 Change “Except for” to “As well as”
- p. 6051, l. 15 For clarity and contrast with the previous sentence, I would suggest: “LOTOS-EURO uses *a fifth system*, a dynamic mixing-layer coordinate system with ...”
- p. 6051, l. 22 500 hPa (~5.5 km) vs. 2.5 to 15 km – is MM5-CHIMERE really an exception?
- p. 6051, l. 27 Section 5.4 does not exist.
- p. 6052, l. 2 Rather than “overviewed”, substitute “reviewed” or “examined”.
- p. 6052, l. 3 Perhaps “A short description of evaluation practices and status for each individual CWF model is presented in Table 10”.
- p. 6052, l. 14 Rather than “singular”, substitute “singly” or “individually”.
- p. 6052, l. 15 “*on* the results”
- p. 6052, l. 16 Perhaps “... techniques include, for instance, *examination of* the role of emissions ...”
- p. 6052, l. 18 Perhaps “In recent years, *results from* an increasing number ...”
- p. 6053, l. 5 Perhaps “a few uncertain *input* parameters”
- p. 6053, l. 10 Perhaps “with respect to all sources and *input* parameters”
- p. 6053, l. 13 “and will include adjoints of both”
- p. 6054, l. 1 A better title for Section 5.2 might be “Single-model evaluation studies” or “Individual-model evaluation studies” given the content of this section and the title of the next section (“Multi-model evaluation studies”).
- p. 6054, l. 3 Replace “critically” with “properly” or “rigorously”.
- p. 6054, l. 26 Suggest “... and (iv) operational *approaches*”.
- p. 6054, l. 28 “*another* possibility is:”?
- p. 6055, l. 22 “... can include a third step, probabilistic evaluation, which ...”
- p. 6055, l. 24 Should clarify here, given the focus of this section, that “ensemble runs” refers to multiple runs with different configurations of the same model”

- p. 6055, l. 29 In the sentence beginning “Sensitivity tests are one of the most common ...”, you could insert a reference to Section 5.1.
- p. 6056, l. 2 Does the sentence beginning “A structured intercomparison among models ...” belong in this section or rather in the next section?
- p. 6056, l. 5 Is it inconsistent to state here that all CWF modelling systems described in this manuscript are based on a NWP model and a CTM (i.e., off-line structure) when it is stated on p. 5994 that Environ-HIRLAM and WRF-Chem are on-line modelling systems?
- p. 6056, l. 9 Replace “in the following:” with “as follows:”
- p. 6056, l. 10 Can you provide a reference for this principle?
- p. 6057, l. 6 “Model *intercomparison* studies also offer the chance ...” might be better.
- p. 6057, l. 8 Perhaps “considered *in this article* have been evaluated individually ...”
- p. 6057, l. 11 Perhaps “*Multi-model* validation against *passive-tracer* field experiments ...”
- p. 6060, l. 25 Perhaps “... different models *that* produce the operational forecasts”?
- p. 6061, l. 1 Insert description of Section 6.3.
- p. 6061, l. 20 “... poor or incomplete documentation”
- p. 6061, l. 8 CMAQ and WRF are also in the public domain. What about Eta, MEMO, Unified Model, and GME?
- p. 6062, l. 10 “model output data”
- p. 6062, l. 12 “set up”
- p. 6062, l. 17 Is it accurate to say that CWF has been “regulated” by a “a number of Directives” or rather has it been “guided” or “influenced”. And whose Directives, i.e., which agency or government or supranational body is responsible?
- p. 6063, l. 14 Replace “oil company distilleries” with “petroleum refineries”.
- p. 6064, l. 7 Perhaps “Environmental decision and policy makers”
- p. 6064, l. 8 Perhaps “These users are responsible for ...”
- p. 6063, l. 19 Do you mean “tenths” or do you mean “tens”?
- p. 6065, l. 20 Rather than “street panels”, perhaps “dynamic street-level displays” or “updated street-level billboards”?
- p. 6066, l. 2 For parallelism with l. 8, perhaps “Those that disseminate air quality (AQ) information based on observational data”?
- p. 6066, l. 14 Would “artificial-intelligence models” be better?
- p. 6066, l. 21 Would “... Portal, which has been implemented in the framework of COST ...” be better?

- p. 6066, l. 25 State how many of 18 modelling systems discussed in this manuscript are included in the 20 systems in the Portal.
- p. 6067, l. 13 What does the acronym “ACT” stand for? Is it defined in the manuscript?
- p. 6067, l. 17 Is “and chemical modelling” really superfluous in the Section 7.1 title since the section text does not discuss chemical modelling at all? The same question is relevant for the “chemical uncertainties” mentioned on l. 12.
- p. 6068, l. 3 Rather than “aerosols”, whose dictionary definition is along the lines of “a suspension of fine solid or liquid particles in gas”, perhaps “PM”.
- p. 6070, l. 14 Replace “approaching” by “approaches”
- p. 6070, l. 16 “Evaluate”, not “validate”
- p. 6070, l. 26 “NWP data *were* rarely available”
- p. 6071, l. 1 Join two sentences: “... for mesoscale air-pollution forecasting, this situation has changed ...”
- p. 6071, l. 10 “environmental forecasting”
- p. 6071, l. 17 Same comment as p. 6068 regarding “aerosols”
- p. 6071, l. 22 “... not the best way for model integration ...”
- p. 6072, l. 20 “*The* recently established new COST Action ...”
- p. 6072, l. 23 “... new generation *of* on-line integrated ...”
- p. 6073, l. 3 Please clarify that these are “*chemical* boundary conditions”.
- p. 6073, l. 10 “with boundary conditions *provided* from larger-scale ...”
- p. 6073, l. 17 “provided” – past tense?
- p. 6073, l. 20 “... global models should in principle improve predictive skill” might better reflect the true state of affairs.
- p. 6074 If “ensemble Kalman filter” is the intended term, it should be used in full and it could be abbreviated after its first use as “EnKF”.
- p. 6075, l. 19 Perhaps “Another challenge for the use of data assimilation in CWF models ...”
- p. 6076, l. 13 Given the discussion in the previous paragraph, would it be better to insert the word(s) “other” or “more sophisticated” before the phrase “data assimilation methods can be applied”?
- p. 6076, l. 28 Perhaps “... is *given* by the ACCENT-TROPSAT-2 report”.
- p. 6077, l. 2 Perhaps “satellite-based *abundance* data, which ...”
- p. 6077, l. 5 “one then has to address the ...”
- p. 6077, l. 13 “At some stage (*of what?*), a data assimilation routine may be used (*for what?*)”?

- p. 6078, l. 21 Could also mention the additional complexity and challenge of representing cloud-topped boundary layers?
- p. 6079, l. 14 What is the “convective parameterization *exercise*”?
- p. 6080, l. 22 “a set” would be better.
- p. 6080, l. 24 “at ground level” and “Clearly, a data comparison”.
- p. 6080, l. 26 “atmosphere”
- p. 6080, l. 26 Being specific is a good thing: for example, “Whenever possible, vertical profiles of air pollutants such as MOZAIC data (ref.) should be ...”
- p. 6081, l. 2 Perhaps delete phrase “or PM measures”
- p. 6081, l. 7 Would be useful to give a relevant supersite reference or two here.
- p. 6082, l. 15 “representative” would probably be better in this context; it would also be helpful to give a relevant incommensurability reference or two here.
- p. 6082, l. 24 Perhaps “... complement to the parameters referred to above”.
- p. 6083, l. 2 Good opportunity here to reference Section 5.3.
- p. 6083, l. 3 AQMEII is “... International Initiative”
- p. 6083, l. 8 “... during the last *few* decades”
- p. 6083, l. 12 Replace “imperfectness” with “imperfection”.
- p. 6083, l. 22 I would suggest “ensemble prediction *in NWP* is beyond the scope”
- p. 6083, l. 28 I would suggest “... to be an area of active research *in NWP*”.
- p. 6084, l. 6 Both A, B, and C (normal usage is “Both A and B”)?
- p. 6084, l. 18 Perhaps “As in NWP, these studies ...”
- p. 6085, l. 5 Perhaps “improvement in CWF performances will be based on the improvement ...”
- p. 6085, l. 7 “hybridizing”? And can you provide a reference?
- p. 6086, l. 17 There needs to be a bridging sentence before the questions “How ...?” and “Moreover, how ...?”.
- p. 6089 For OPANA, should first word be “operational”? Add NOAA to list?
- p. 6090 Missing references include
- Frohn and Brandt (2006); see p. 6008
  - Janic (1994); see p. 6016
  - Rötzer and Chmielewski (2001); see p. 6048
  - Siljamo et al. (2008); see p. 6006
  - Zhu et al. (2009); see p. 6065



- p. 6097, l. 3 Reference is out of order.
- p. 6104, l. 18 “dimethyl sulphide”?
- p. 6109, l. 11 Number of pages?
- p. 6115, l. 13 I think the co-authors should be Phillips, Sarwar, and Jang.
- p. 6120, l. 26 “parameterization”
- p. 6123, l. 17 It would be more appropriate to give the reference for the revised English language version (1976, Hermosa Publishers, Albuquerque, New Mexico, 453 pp. [ISBN 0-913478-05-9])
- p. 6126, l. 9 Current status?
- p. 6129, l. 10 “Institut für Meteorologie”?
- p. 6136, l. 9 “fremlagt ved” – English translation?
- p. 6145 In Table 4, rather than “ND”, would “NC” (“not considered”) or “NI” (“not included”) be a better description?
- p. 6146 For Gross and Stockwell (2003), should RACM go in the “Other” column?
- p. 6148 In Table 7, CHIMERE and CMAQ are not listed in alphabetical order.
- p. 6152, l. 1 Suggest “The particulate *natural* emissions ...”
- p. 6158 “publicly available”
- p. 6161 Since STEM is not included in Table 13, the table title should be modified to qualify which models are considered.