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## ***Interactive comment on “Online coupled regional meteorology-chemistry models in Europe: current status and prospects” by A. Baklanov et al.***

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Overall I think that the paper should be published in ACP, subject to revisions which should take less than a few weeks to carry out. My comments are generally requests for clarification, as opposed to serious issues with the descriptions, as well as identifying places where the authors are repeating things said elsewhere in the paper or which are not necessarily of key relevance for the topic under review.

One key concern I have is the length of the paper, which is much longer than is usually acceptable for a journal publication (it would make a very good reference book!). I've included some suggestions on how the flow and information presentation could be improved with reduction of some sections and/or moving these to an appendix..

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## Specific comments:

Page 12546, line 14: "NWP does not require detailed chemical processes". This statement presupposes that the impact of feedbacks on NWP is small, while other parts of the paper note that long term evaluation of full feedback models is still needed (and will hopefully come to light under the AQMEII-2 project). It would be more accurate to state that "NWP in its current state does not include detailed chemical processes"

Page 12546, line 20-21, "For CWF ... on-line integration definitely improves AQ..." This statement has been made in advance of the review itself – the authors have not made a case for these improvements at this stage in the paper. Some references that support the statement are needed here, or a better justification for the statement.

Page 12547, last paragraph of section 1: At the same time, the authors acknowledge elsewhere that longer evaluation periods are needed in order to determine the impacts of on-line feedbacks on the meteorological forecast and vice-versa. This should also be mentioned here: the time periods which have been used for evaluation (up to this point, in the literature) have been short – and this may also affect the ratings of which aspects are more or less relevant. So a caveat is needed to that effect in the manuscript.

Page 12547, line 26, days to decades: consider the impact of feedbacks associated with convective cloud formation (indirect effect) via CCN growth. This may be a relatively fast process (hours rather than days for the time scale).

Page 1258, line 18 – Tables 1 and 2 are missing the impact of meteorology on the removal of chemicals through deposition (i.e. consider the met-dependent portions of surface resistance and deposition velocity calculations). Note also that geographical input data such as vegetation type and land use type are key inputs; not sure where this fits in to the tables. Table 2: note that the introduction of aerosols may affect wind direction as well as speed.

Page 12549, line 10: Connection between aerosol absorption of sunlight and cloud

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liquid water is unclear to me. I think there may be a few processes that are needed between the two to make that connection clear.

Page 12550, discussion on Table 3: Table 3 is interesting, but also shows biases, in that it is dependent on the expertise and background of the respondents. The authors note (line 21) that the results might be affected by individual opinions – definitely, and (line 25) that very few climate modellers were involved in the survey. There needs to be a few lines of justification regarding why a survey that is subjective in this nature should be done.

Page 12554, line 20-21. ABL heights – explain the importance of the ABL (as opposed to, for example, the values of diffusion constants – is the ABL a useful diagnostic of model performance or a variable required for prediction)?

Page 12556, line 17-19. It may be worth referencing mechanism intercomparison papers here (e.g. Kuhn et al 1999): the level of detail in a chemical mechanism does not necessarily imply an increased level of accuracy in the simulated results; the authors state that computational constraints are the only thing preventing use of a very complex mechanism – what evidence is available that suggests the very detailed mechanisms actually do a significantly better job in the context of 3D AQ models? It would be good to have something quantitative.

Page 12556, lines 21-27: This discussion implies that the gas-phase mechanism is the key issue in accurately representing secondary organic aerosol formation. However, there is good evidence to suggest that significant SOA formation may occur in association with particle surface reactions, cloud chemistry, etc. This should be mentioned here. It would be good to reference Gong et al (2011) here in that context as well as in the latter discussion in the manuscript.

Page 12559, lines 1-6: Note that each new mechanism requires a new set of emissions data, due to the VOC speciation changing between mechanisms. Also, the connections to other processes (e.g. deposition) will be species, hence mechanism dependant. The

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point being that changing the mechanism in the gas-phase portion of the model is only part of the setup required for switching mechanisms in a 3D model.

Page 12559, lines 8 to 20: There are a number of statements being made in this paragraph which need some more justification and/or references. “NWP does not depend on detailed chemical processes that may be necessary to predict air quality” – in the absence of long-time period simulations with and without feedbacks in a fully coupled model, how can this statement be made? Certainly current NWP models do not include detailed chemistry, but the statement reads as if for NWP, the feedbacks are not important. Has this been proven? “Needs just enough complexity to be able to model aerosol effects on radiative and precipitative processes” – how much complexity is that? Determining the extent to which NWP may be improved through coupled models with feedbacks is an ongoing and interesting topic of current research – I think that the “jury is still out” with regards to their importance. So these statements are perhaps premature.

Page 12559, last paragraph: yes, a central database for gas-phase mechanisms would be helpful – it would also be helpful to have methodologies for SOA formation be a part of the same or associated database.

Page 12563, top paragraph describing inorganic heterogeneous chemistry approaches. It is important to note somewhere in the text that these modules (even when solving sub-spaces of the problem as in the case of ISORROPIA-1,-II) usually are used in “bulk mode”, that is, inorganic aerosol mass across the size distribution is added up within each species, and partitioning is with respect to this bulk mass. This is done for computational efficiency, rather than solving local problems at different locations within the size distribution. Also page 12565, line 21: “is in a bulk thermodynamic equilibrium” is more precise.

Pages 12563, 12564, discussion on SOA formation. It should be noted that all three of these approaches tend to assume that all of the SOA formation results from gas-phase

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reactions, while recent laboratory evidence is starting to suggest that other mechanisms (e.g. surface reactions, cloud chemistry, acid catalysis of organic polymer formation) may be required to adequately represent the formation of organic aerosols.

Page 12565, line 14 – would also be good to reference Gong et al 2011 here as a review on this topic.

Page 12574, line 13-15: how many AQ models explicitly model Fe, Al, and PAH compounds? Is this based on observations or on models which have evaluated these impacts?

Page 12574, section 4.5.1: might be better to title this “Direct radiative effects of gases and aerosols”, since section does not deal with aerosol indirect effect.

Page 12576, last sentence of 4.5.2: What data is available for this validation? Also, what data are available on fundamental aerosol properties such as complex refractive index as a function of chemical speciation within the aerosol? The section does not mention issues regarding internally mixed, versus core/shell, versus multiple core/shell approaches to working out the impact of aerosols (recent work by Binkowski and by Jacobson). It would be useful/interesting to compare the different complex refractive index values used in the different models in a table.

Section 4.7.1, second two paragraphs. One aspect of the “natural” emissions is that no matter how good the emissions algorithm is, the model will be dependent on the accuracy of the underlying input data, which in this case is land use type, vegetation type, etc. Given that this is the case, the authors should comment on the availability and accuracy of that data, and whether any improvements are required to improve on-line model accuracy. Also, many of the wind-blown dust processes make assumptions regarding soil type and soil moisture – parameters that may be available in meteorological model input databases, and in any event a point for further coupling between the meteorology and chemistry.

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Page 12581, final paragraph of 4.7.1: note that on the local or urban scale the contribution of anthropogenic VOC emissions may be quite significant, since those emissions occur over a very limited spatial domain. Consequently, their impact at the locations where most of the human population resides may be quite large. Referencing just the large-scale impact may thus underestimate the local impact (which may be of greater importance from the standpoint of policy implications of the model results).

Page 12582, line 2: Suggest “only possible pathways to remove” should be “only possible pathways (aside from chemical transformation) to remove”.

Page 12591, line 18: need to define what is meant by a “coupler” in this context.

Page 12591, line 27: “that exchange information” The word “exchange” implies a two-way interaction, while the sentence is in reference to offline models, where the information passing is one-way, and via input/output files. Clarify.

Page 12592: The authors introduce “slightly coupled” models here for the first time. What does this mean – how is this defined? It would be better to stick with the earlier terminology and/or define this in the earlier section where off and on-line models are defined.

Page 12593, last sentence of section 5.2. Computational demands also rapidly increase with off-line models; this is not something that is off or on-line specific, so the statement can be removed.

Page 12598, statements on bottom of page of the transient nature of initial conditions. While the initial conditions may be transient, the use of data assimilation to improve boundary conditions in a regional run may be quite important. Consider the findings of the HTAP experiment, wherein the impact of emissions changes in different continents were evaluated based on global-model-predicted changes in that and other continents. One of the important findings there was that outside of the summer season, long range transport of ozone from one continent to another could have a significant

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impact on ozone levels in the destination continent. Following this concept – if data assimilation is used to create better ozone boundary conditions for a regional model run, it could have a substantial impact on regional model predictions. Perhaps the discussion could be linked to 5.5.3 at this point?

Page 12600, line 23: “simulated online” versus “two-way coupled”? So is the first supposed to be with reference to online but not coupled modelling? Unclear. Similarly, page 12601, line 1 – online configuration versus current coupled system? Unclear. Also, more efficient with respect to what, data assimilation? Needs to be reworded.

Page 12601, line 21-23: The sentence starting with “Nevertheless” doesn’t make sense: what is probably the easiest CDA technique to implement? Something has been inadvertently deleted, here. Line 27: need to explain what is meant by parameter estimation in the context of online models.

Section 6.1: This describes past use of online coupled models, but its not clear what results from the studies were unique as the result of the use of that type of model as opposed to off-line models. A key question that I kept asking while reading this section was, “What was the unique contribution of the on-line approach”? This section would be better removed, or shortened and perhaps added to the introduction as background for on-line modelling work in Europe. The last few lines (12604, line 23 and down) talk specifically of how the on-line approach improved the results – that’s what should be the focus of this section, if possible.

Section 6.2: it may be the way the information was reported here, but there seems to be a significant lack of comparison with observations in the modelling studies reported here. That is, only a few studies are mentioned as having had an impact (positive or negative) on the accuracy of the model predictions with respect to observations. Is that because most of the on-line work in Europe has been sensitivity studies to date, or is that information available in the papers quoted? If the latter, it should be summarized as part of this section. If the former, then the need for detailed evaluation should be

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mentioned as one of the outcomes of this section.

Page 12606, line 20, also page 12608, lines 7-9: for these quantitative statements of impact, mention the significance relative to, e.g. typical radiative budget. i.e. what can be said regarding the relative impact of the feedbacks compared to other factors? Are the feedbacks capable of having a significant effect, are they lost in the noise, etc.? For direct aerosol effect impact being “substantial” – can this be quantified? Were these model sensitivity tests, or was there any attempt in the studies to rate the impact relative to observations (i.e. any improvement via feedbacks)?

Page 12607, line 1 – does this imply that the impact of feedbacks on turbulence had a minor effect on the predicted model results?

Page 12608, lines 18 to 24: the impact on model results is described, but not whether the model results improved (either relative to observations or from a theoretical standpoint). Do these studies make the case that on-line coupled models are “doing a better job” or is there insufficient evaluation at this point to say this? Page 12609, lines 10 to 15: this is the sort of thing that should be emphasized from the other studies as well: the on-line model in this case clearly has at least the potential to correct an existing deficiency in modelling.

Page 12609, line 19: brightening of what? Cloud top albedo? Surface albedo? Clarify.

Redundancies in the manuscript (things that could be removed or reduced):

Page 12544, first paragraph of the introduction repeats information already in the abstract – remove.

Page 12583, lines 10 through 20: this information has appeared earlier in the manuscript and does not need to be included here. In that respect, anything that has been included in previous sections need not be repeated here – it would be sufficient to start this section at “Several additional feedback mechanisms” (page 12584, line 7).

Page 12588, section 5.1: This is a good summary of the state of the science for nu-  
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merical methods used for modelling – but it is not specific to the topic of the paper; the on-line aspects of modelling. This section should be greatly shortened, removed, or put in an appendix. The latter might be the best route – it's a good summary, but it detracts from the paper's main focus in its current location.

Page 12593, section 5.3. As in section 5.1, this section deals with generic issues that are not specific to on-line models per se, but any model configuration. As such, it is not part of the main topic of the paper. The section should be placed in an appendix or removed altogether (with my preference being an appendix – the information is useful, but not as relevant to the main topic under discussion). Page 12594, last 8 lines: the discussion does not explain why CAF represents a better means of coding – what is the advantage, and why?

Page 12595, section 5.4. Again, these issues are common to both off-line and on-line models, and should be removed or placed in an appendix. One issue that I would think would be worth mentioning in the main body of the paper would be the extent to which boundary conditions provided from a global on-line model may be superior to those from a global off-line model, if any work has been done on that topic. The last two paragraphs of the section lists the different approaches used, but doesn't interpret which methods are better / worse, which would be of interest to the broader community.

Page 12610, section 6.3: Model evaluation is a very important topic, but it is not a topic that is specific to the class of on-line models. A large part of this section could be removed or placed in an appendix – the section could easily start at line 3 page 12614.

Page 12614, line 19: why are new and improved strategies required for online model evaluation? The reasoning for this statement doesn't become clear until the last paragraph, and should be presented earlier. Line 23 "In many cases..." this presupposes that the differences due to feedbacks are small. Are they? What evidence is in the literature to this effect? Maybe include a few references. Line 26: should that be "much data assimilation", not "data simulation"?

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Page 12626, line 5: I don't recall these issues being discussed in the paper (ship emissions, aviation - why/how has this become a recommendation? Line 23: "thus should be aimed at."? Reword this, unclear.

Appendix A: the first part of the descriptions should focus on the degree of coupling in place. This is not clear from the descriptions as they currently stand.

Minor issues:

Page 12544, line 4: replace "attempt reviewing" with "review"

Page 12546, paragraph lines 13 through 22, as a point of interest (and for potential inclusion here), the on-line model GEM-MACH has been used for operational air-quality forecasting at Environment Canada, providing North American forecasts of O3, PM2.5, and NO2 to the public since 2009 (Moran et al, 2010). Also, page 12552, lines 15-20, please add the same reference here.

Page 12555, line 4: "calculate fluxes" should become "calculate radiative fluxes"

Page 12561, line 9: "deviations variable" should be "deviations of variables"

Page 12561, line 20: GEM-MACH model, Moran et al, uses 2 bins for operational configuration, 12 for research, FYI.

Page 12566, line 15: "degree" should be "degrees"

Page 12568, line 19: "It depends" – What depends? Not clear.

Page 12568, section 4.5.1 title: perhaps this should be "Aerosol-cloud interactions in online models with diagnostic property equations"; it seems to describe the methods used in on-line models which do not include feedbacks. Line 17: the "above example" mentioned: these sorts of parameterizations are unnecessary if schemes such as Abdul-Razzak and Ghan, 2002 are used.

Page 12572, line 27: "opposite is true" Unclear – i.e. concentration is high enough for

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global impact, or the temperature for ice nucleation for BC & OC is relatively low?

Page 12621, line 1: “consistency” should be “mass consistency”?

References:

Moran MD, Ménard S, Talbot D, Huang P, Makar PA, Gong W, Landry H, Gravel S, Gong S, Crevier L-P, Kallaur A, Sassi M (2010) Particulate-matter forecasting with GEM-MACH15, a new Canadian air-quality forecast model. In: *Air Pollution Modelling and Its Application XX*, Steyn DG, Rao ST (eds), Springer, Dordrecht, 289-292.

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