

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

Anonymous Referee #3

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General Comments:

This paper is a comprehensive overview of online and coupled meteorology-chemistry models that are currently in use in Europe. While the stated goal is to focus on integrated meteorology-chemistry models, the paper provides an extensive description of the chemical and physical processes involved in chemical transport modeling (section 4). Although it can be argued that this is important background for the paper's purpose, it seems disproportionate compared to the description of the processes and modeling techniques unique to online models such as aerosol direct and indirect radiative feedback effects. It seems like more than half of the paper is describing aspects of AQ modeling, including extensive discussion of meteorologically dependent emissions that are equally germane to offline models. Thus, I think these sections could be greatly reduced and instead have more in-depth description and discussion of processes that are only possible in online or coupled systems. While there is a complete survey of the techniques used for direct and indirect aerosol feedbacks there is not enough explanation of how these models work, especially the CCN activation algorithms. It would be more enlightening if the paper could give some judgement as to which techniques are most realistic and the trade-offs in terms of computational cost. As explained, evaluation of indirect effects is difficult but on theoretical basis some techniques must be more valid than others.

Thank you for reviewing our manuscript and providing valuable comments. We have incorporated all your comments into our revised version. Please find all our replies in red colour, following your remarks, which we copied and kept in black.

Section 4:

We agree about the lack of uniformity and that the level of detail is different in different sections. More specifically, the different subjects touched upon in our manuscript have different levels of scientific understanding, and are of different relevance for online coupled models. This is the reason, why the level of details varies from section to section. To address your comments, we've removed some minor details in this section, and also streamlined and harmonized the sections in the revised manuscript.

Please see below our point-by-point replies to the remaining general comments and specific comments.

There would seem to be more opportunities for integration of meteorological and chemical processes than are discussed in this paper. In particular, the interactions between chemistry and clouds could go much beyond the effects of aerosols as CCN. For example, cloud droplet size spectra could be used in the aqueous chemistry calculations rather than a bulk cloud water approach that is typically used in offline models. Also, once a portion of the aerosol size spectra are activated the partitioning of the aerosol into cloud droplets and interstitial aerosol could be fed back to the chemical model where aerosol and aqueous process would be affected. In addition, aqueous chemical species could be advected through the model grid along with microphysical species.

While some of these processes may not be worth the additional complexity and computational cost, they probably should be mentioned in the paper.

We agree with the reviewer that there are several processes that are currently not taken into account in the online coupled models and some of them might be important. One example is the release of aged aerosol when cloud droplets are evaporated. However, little is known about the aerosol composition at this stage, thus we did not discuss it. As our paper is already quite long we decided to concentrate on those processes that are currently treated in online coupled models used in Europe.

The terminology for coupled meteorology-chemistry models is confusing with inconsistencies throughout the paper. Section 3 attempts to define the distinctions between “online access models” and “online integrated models”. However, in the same section the term “online coupled models” is used with no explanation of how this label relates to the other terms. Furthermore, in section 3.2 it is stated that online access models have meteorological and chemical data available at each timestep but later this is contradicted (page 12616): “: : some models (e.g. RACMO2/LOTOS-EUROS, COSMOMUSCAT) follow the online access approach, with a data exchange between the chemistry and meteorology modules not on each model time step.” It is important to make the terminology clear and consistent.

We agree that in the submitted paper the wording was not always clear. The terminology for coupled meteorology chemistry models has been defined and used consistently throughout the text in the revised manuscript.

The quality of the English is rather uneven. Some parts are well written while others are poorly written. Also, some sections are composed of long paragraphs while others often have very brief paragraphs. A greater effort should be made to improve the writing and harmonize the style.

The raised issues have been addressed with a more consistent writing style, better presentation of the material, and improved readability throughout the text in the revised manuscript. The English is checked by native speakers.

Overall, this paper is a valuable addition to the literature that provides a comprehensive description of all the major components of coupled meteorology-chemistry models with particular emphasis on integrated models and the chemistry to meteorology feedbacks that are only possible in such online systems. It is also quite interesting to see how the variety of models compare to each other and both the many similarities and differences in their approaches to the same processes. Once the general and specific issues mentioned here are dealt with, this should be acceptable for publication in ACP.

Specific comments:

P12545ln6: OCMC should be spelled out the first time it's used.

OCMC is the acronym for “online coupled meteorology chemistry”, which has been defined in the revised manuscript not only in Table 9 (acronyms) but also in the text..

P12556ln5-6: This is one of the places where the interactions of aerosols and cloud droplets could be taken a step further. It is stated that aqueous chemistry is a function of liquid water content but in coupled models it could also take droplet size spectra into account.

This point has been added in the revised manuscript.

P12556ln6-9: This sentence is poorly written. The statement that radiatively active gases are important for climate while aerosols are important for meteorology is incomplete since aerosols are also important for climate, especially at regional scales.

This sentence is rewritten.

P12557ln23: There is also a CB6

This has been included in text:

“Update the Carbon-Bond chemical mechanism with new aromatic, isoprene, and inorganic reactions (CB6).”

P12561In2-13: It is stated that a 3-moment modal approach could be problematic because of inconsistencies due to advection. But then it is stated that holding the standard deviation constant may lead to large errors. Examples of models using both approaches are mentioned but with no support or elaboration for either assertion. The reader is left with no insight into which approach is preferred or the tradeoffs involved.

We agree with the reviewer that we gave the impression that using a 2 moment approach leads to unacceptable errors. We therefore removed ‘large errors’ by errors. The fact that different approaches are currently used by the scientific community shows that there is no perfect approach. However, the paper by Zang et al. (2009) gives the pros and cons of the different methods.

P12562 Sections 4.3.4 and 4.3.5: These sections seem ridiculously brief. I suggest combining them into one section perhaps entitled “Aerosol mixing state”. In addition to internal and external the core-shell and multiple inclusion models for BC should be discussed. However, since aerosol aging and the coating of BC particles is discussed in section 4.3.7, it may be better to move these sections to be part of that section. I think this whole larger section (4.3.x) should be reorganized and re-written for more logical progression and improved English and more uniform style (i.e. some sections are long and others are short).

We agree with the reviewer, who is supported by other reviewers of our manuscript. We therefore rearranged section 4.3 to make it more logical and to reduce the number of subsections.

P12562In 14-15: This sentence seems to contradict the earlier sentence at lines 2-3.

Lines 14-15 stated that a “full discretization of both size and chemical composition... has not yet been incorporated into 3-D models”, which is actually consistent with lines 2-3 that stated “It is impossible to describe the full complexity of aerosol chemical composition in 3-D numerical models.” Note that both statements indicate that a full discretization of chemical composition is not yet possible in 3-D models. To avoid confusion, we modified the lines 14-15 to be:

“A full discretization of both size and chemical composition is feasible in box models (Dergaoui et al., 2013), but it has not yet been incorporated into 3-D models.”

P12564In5-6: The VBS model is described as including “chemical aging within the particle” when I think the VBS concept is that chemical aging occurs via gas-phase reactions.

The reviewer is right, we corrected the text.

P12564 Section4.3.6: Another SOA process is the formation of non-volatile SOA via heterogeneous uptake and particle phase reaction.

We added this to the text and give references.

P12565 Section4.3.9: This section is redundant and should be merged with section 4.3.6. More reason for reorganization of this whole section.

We agree and merged both sections.

P12567In20-22: This sentence seems to contradict the later discussion of CCN activation as function of supersaturation.

P12568In24-25: This statement that “all online models have cloud schemes that to some extent represent the effects of aerosols on cloud” is clearly not correct.

Answer to p12567/12658 comments: What we meant here is that even if one prescribed a CCN concentration that differs over land and ocean, to some extent, such a prescription, though highly simplified, represents very crudely the influence of aerosols. The sentence has been rewritten in the text and is given below:

“many online models have cloud schemes that either crudely or more accurately represent the effects of aerosols on cloud”.

P12570In3: I think they meant to say: “In all such models: : :”

Yes, this is what we meant.

P12572In27: What is meant by: “The opposite is true: : :”?

This has been rewritten to:

“Black carbon and carbonaceous particles, on the other hand, are much more numerous, but it is not yet clear, if they nucleate ice well above the homogeneous freezing temperature.”

P12574Ins15-20: This paragraph is confusing. Also, where are the effects of radiatively absorbing aerosols, such as BC, inside cloud droplets discussed? The heating caused by in-droplet BC radiation absorption can cause evaporation of the droplets.

The paragraph has been rephrased. Effects of absorbing aerosols are discussed in the references added at the end of the paragraph, while existing implementations of droplet dynamics are reviewed in section 4.5.2.

P12580In11-13: References for the ammonia bidirectional flux scheme in WRF-CMAQ should be included (Cooter et al 2012, Biogeosciences; Bash et al 2013, Biogeosciences; Pleim et al 2013, JGR)

The suggested references have been added.

P12582In7-11: Dry deposition also depends on the land use and vegetation characteristics and the chemical and physical properties of gasses. The list of dry deposition references might also include Pleim and Ran Atmosphere 2011, 2, 271-302; doi:10.3390/atmos2030271.

This point has been added along with references in the revised manuscript.

P12583In10-18: This is almost exactly repeated from Section 2. Why?

This section has been rewritten and repetition has been removed in the revised manuscript.

P12583In21-27: This paragraph implies that only BC aerosols can cause the effects of lower PBL heights and higher concentrations. While BC is particularly able to warm upper levels of the PBL by radiation absorption, all aerosols reduce SW radiation at the ground thereby reducing surface temperatures leading to more stable and shallower PBLs.

Those statements were made in the context of Zhang et al. (2010a), in which “the cooling at the surface resulting from reduced solar radiation” was indeed by all aerosols. To avoid confusion, the statements have been revised in the main text (see below):

“Zhang et al. (2010a) analysed with the online model WRF-Chem such a “chain effect” over the continental U.S. and simulated enhanced stability as a result of the warming caused by BC in the ABL and the cooling at the surface that results from reduced solar radiation by all aerosols. This effect in turn further increases air pollution concentrations over areas, where air pollution is already severe.”

P12584In1-6: In addition to being poorly written, this paragraph overstates the inferiority of so-called online access models in simulating the chain effects.

The relevant statements have been revised.

P12584In7-11: This is a badly written and useless paragraph. Section 5.1: Shouldn't positive definiteness and monotonicity also be mentioned as desirable properties for advection schemes?

To make it more obvious that this is mentioned we add “(positive definiteness, monotonicity etc.)” after the word “shape-preservation” in line 26 on page 12587. The subject is dealt with in section 5.1.2. The section has been re-formulated to make this more obvious (and for other reasons).

Section 5.1.1: The terminology here is confusing. What is meant by volume density? It appears from the equations that this is plain density, i.e. mass per volume. Is mixing ratio mass mixing ratio?
We agree that “density” should be used for the mass per volume and “concentration” should be used for mass per mass (i.e. mixing ratio). This has been used in the revised version.

Equation 2 seems to be incorrect. Shouldn't the RHS be $-V_{del}(q_i)$?
This typo has been corrected in Eq. 2 in section 5.1.1.

P12590ln13-15: It should be mentioned that these statements are about horizontal turbulent mixing and not vertical turbulent mixing. Also, numerical diffusion may not always be much greater than turbulent diffusion, especially at finer horizontal grid resolution.
This point has been incorporated into the revised manuscript.

P12591ln25-28: This sentence needs to be rewritten for more clarity. It mentions offline models but then also says that information is exchanged through a coupler, which sounds more like an online access model. The next sentence says it's one-way. This is very confusing.
The sentence has been reworded, and the use of coupler is avoided for clarity.

P12614ln26: “simulation” should be “assimilation”.
The second part of the sentence that contains “simulation” was removed to avoid confusion.

P12624ln 12-14: This statement is overstated. Not only online integrated models can include aerosol dynamics and feedbacks but also online access models.
The sentence has been rewritten.

P12625ln25-27: This sentence is poor grammar and incorrect. Online access models are not necessarily more expensive. In fact, they can be much more computationally efficient since the CTM can generally be integrated at a longer time timestep than the meteorology model.
We agree that there are some advantages of online access models as well, but in general they have some serious limitations compared to online integrated models. Relevant statements about online access models have been modified correspondingly.

P12627ln25: “base” should be “basis”
The suggested change has been made.

Table 5: The WRF model has far more convective, PBL, and radiation options than listed here. For PBL the YSU scheme is most commonly used for WRF-Chem and the ACM2 scheme is used for WRF-CMAQ. For radiation, the CAM, Goddard, and RRTMG schemes are the only ones capable of aerosol direct feedbacks using WRF-Chem and WRF-CMAQ.
Table5 has been modified with more options for WRF convection, PBL, and radiation schemes. Due to limited space, not all the schemes are listed in this table.