

Integrated Meteorology Chemistry Models: Challenges, gaps, needs and future directions

White Paper *Draft 1*

Abstract

Online coupled meteorology atmospheric chemistry models have undergone a rapid evolution in recent years. Although mainly developed by the air quality modelling community, these models are also of interest for numerical weather prediction and climate modelling as they can consider not only the effects of meteorology on air quality, but also the potentially important effects of atmospheric composition on weather. Two ways of online coupling can be distinguished: online integrated and online access coupling. Online integrated models simulate meteorology and chemistry over the same grid in one model using one main timestep for integration. Online access models use independent meteorology and chemistry modules that might even have different grids, but exchange meteorology and chemistry data on a regular and frequent basis. This paper offers a review of the current research status of online coupled meteorology and atmospheric chemistry modelling, a survey of processes relevant to the interactions between atmospheric physics, dynamics and composition; and highlights selected scientific issues and emerging challenges that require proper consideration to improve the reliability and usability of these models for the three scientific communities: air quality, numerical meteorology modelling (including weather prediction) and climate modelling. It presents a synthesis of scientific progress and provides recommendations for future research directions and priorities in the development, application and evaluation of online coupled models.

1. Introduction into coupled meteorology-chemistry modelling

Coupling of atmospheric dynamics, pollutant transport, chemical reactions and atmospheric composition for modelling environmental impacts, climate change, weather forecasts and air quality will remain one of the most challenging tasks over the next decades as they all involve strongly integrated processes. It is well accepted that weather has a profound impact on air quality (AQ) and atmospheric transport of hazardous materials. It is also recognised that atmospheric composition can influence both weather and climate directly by changing the atmospheric radiation budget or indirectly by affecting cloud formation and precipitation. Until recently however, because of the scientific complexities and lack of computational power, atmospheric chemistry and weather forecasting have developed as separate disciplines, leading to the development of separate modelling systems that are only loosely coupled. This is particularly true for regional scale models, whereas for global scale and in particular stratospheric modelling, the development and availability of online coupled models is more advanced.

The dramatic increase in computer power during the last decade enables us to use high spatial resolutions (e.g. < a few km) in numerical weather prediction (NWP) and meteorological

¹ Based mostly on the EuMetChem paper: Baklanov, A., Schlünzen, K., Suppan, P., Baldasano, J., Brunner, D., Aksoyoglu, S., Carmichael, G., Douros, J., Flemming, J., Forkel, R., Galmarini, S., Gauss, M., Grell, G., Hirtl, M., Joffre, S., Jorba, O., Kaas, E., Kaasik, M., Kallos, G., Kong, X., Korsholm, U., Kurganskiy, A., Kushta, J., Lohmann, U., Mahura, A., Manders-Groot, A., Maurizi, A., Moussiopoulos, N., Rao, S. T., Savage, N., Seigneur, C., Sokhi, R. S., Solazzo, E., Solomos, S., Sørensen, B., Tsegas, G., Vignati, E., Vogel, B., and Zhang, Y.: Online coupled regional meteorology chemistry models in Europe: current status and prospects, *Atmos. Chem. Phys.*, 14, 317-398, doi:10.5194/acp-14-317-2014, 2014.

modelling. Fronts, convective systems, local wind systems, and clouds are being resolved or partly resolved. Furthermore, the complexity of the parametrisation schemes in the models has increased as more and more processes are considered. Additionally, this increased computing capacity can be used for closer coupling of meteorological models (MetM) with atmospheric chemical transport models (CTM) either offline or online. **Offline modelling** implies that the CTM is run after the meteorological simulation is completed, while **online modelling** allows coupling and integration of some of the physical and chemical components to various degrees.

In recognition of the rapid development of coupled meteorology and chemistry modelling, Action ES1004 (EuMetChem) in the European Cooperation in Science and Technology (COST) Framework was launched in February 2011 to develop a European strategy for online integrated air quality (AQ) and meteorology modelling (see the web-site: www.eumetchem.info). The Action does not aim at determining or designing one best model, but to identify and review the main processes and to specify optimal modular structures for online Meteorology Chemistry (MetChem) models to simulate specific atmospheric processes. Furthermore, the COST Action develops recommendations for efficient interfacing and integration of new modules, keeping in mind that there is no one best model, but that the use of an ensemble of models is likely to provide the most skilful simulations (Baklanov et al., 2014). The WMO Working Group for Numerical Experimentation (WGNE) recently initiated also a specific online coupled modelling case study on Aerosol Effects on Numerical Weather Prediction (see the web-site: www.wmo.int/wgne/).

These coupled models are distinguished with respect to the extent of online coupling: online integrated and online access coupling. **Online integrated** meteorology chemistry models handle meteorology and chemistry using the same grid in one model and using one main timestep for integration. **Online access** models use independent meteorology and chemistry models that might even be using different grids, but exchange information from meteorology to chemistry and back to meteorology on a regular and frequent basis. The frequency of data exchange needs to increase as the time scale of the relevant processes becomes smaller. In contrast to online access models, offline models do not exchange data, but merely provide, e.g. meteorology information to drive the chemistry model. The ultimate stage is the online integration of CTM and MetM to produce a unified modelling system with consistent treatments of processes such as advection, turbulence and radiation for both meteorological and chemical quantities. Such an integration allows online integrated meteorology chemistry simulations with two-way interactions (also referred to as feedbacks). Climate modelling is also expanding its capability through the use of an earth system modelling approach that integrates the atmosphere, hydrosphere (including both fresh water and oceans) and biosphere with high spatial and temporal resolution. Climate modelling, however, does not require the implementation of near-real-time data assimilation, which is crucial for the skill of NWP and can also help improve AQ forecasts.

For performing a simulation, the input data need to be tailored to the specific requirements of the atmospheric model. For this purpose several programs are employed for each model, that pre-process data, e.g. meteorology measurements on model grids as initial data, land use data consistent with the model land use categories and emission data in agreement with the used chemical mechanism. Specific programs are also needed for providing output data from an atmospheric model.

Combining two modelling systems for operational applications, each of which have high CPU time and memory requirements, still poses many problems in practice and thus is not

always feasible at NWP or chemical weather forecasting centres. Nevertheless, one can argue that such gradual migration towards ever stronger online coupling of CTMs with MetMs poses a challenging but attractive perspective from the scientific point of view for the sake of both high-quality meteorological and chemical weather forecasting (CWF). While NWP centres, as well as entities responsible for AQ forecasting, are only beginning to discuss whether an online approach is important enough to justify the extra cost (Baklanov, 2010; Grell and Baklanov, 2011; Kukkonen et al., 2012; Zhang et al., 2012a,b), the online integrated approach is already used in many research atmospheric models.

For NWP / CWF centres, an additional benefit of the online approach would be its possible application for meteorological data assimilation (Hollingsworth et al., 2008). This assumes that the modelling system can outperform pollutant concentration climatologies when forecasting concentrations of aerosols and radiatively active gases. The retrieval of satellite data and direct assimilation of radiances is likely to improve both weather and chemical weather forecasts.

Online coupled meteorology and chemistry models have been developed in recent years, particularly in the United States (U.S.) (e.g. Zhang, 2008) and these models are becoming increasingly popular in Europe. Historically, Europe has not adopted a community approach to modelling and this has led to a large number of model development programmes, usually working almost independently, thereby yielding results tailored for specific applications (Baklanov et al., 2011). However, a strategic framework could help to provide a common goal and direction to European research in this field, while still having various models as part of a European model ensemble. The task is manifold since it requires scientific knowledge and practical experience in Met and AQ modelling and forecasting, numerical analysis, atmospheric physics, chemistry and data assimilation.

The focus on integrated systems is timely, since recent research has shown that meteorology and chemistry feedbacks are important in the context of many research areas and applications, including NWP and AQ forecasting, as well as climate and Earth system modelling. However, the relative importance of online integration and of the priorities, requirements, and level of details necessary for representing different processes can vary greatly between applications. Under these circumstances tailored solutions may be required for the three communities: (i) AQ forecasting and assessments, (ii) NWP and Met modelling, (iii) climate and earth system modelling.

For example, current NWP models do not incorporate detailed chemical processes, even though aerosols – via radiative and microphysical processes, can affect fog formation, visibility and precipitation, and thus forecasting skill. For climate modelling, feedbacks from greenhouse gases (GHGs) and aerosols are extremely important, though in most cases (e.g. for long-lived GHGs), online integration of full scale chemistry and aerosol dynamics is not critically needed. For CWF and prediction of atmospheric composition in a changing climate, online integration is expected to improve AQ and atmospheric chemical composition simulations and projections (e.g., Moran et al., 2010). The AQ, Met and climate modelling communities have different targets with respect to temporal and spatial scales, as well as to the processes involved in such modelling. For AQ forecasting, the key issue is usually the ground-level concentrations of pollutants, whereas for weather and climate models, skill is typically based on screen level temperature, precipitation and wind. Since short-lived pollutants influence climate and air quality conditions, the AQ community is interested in online modelling to understand the feedback

mechanisms and to design air quality policies that can maintain future air quality at acceptable levels under changing climate conditions (Alapaty et al., 2011).

Several applications are likely to benefit from online modelling, although they do not clearly belong to one of these three main communities mentioned above. These include bioweather forecasting, pollen warnings, forecasting of hazardous plumes from volcanic eruptions, forest fires, oil and gas fires, dust storms, assessment of methods in geoengineering that involve changes in the radiation balance (e.g. input of sulfate aerosols, artificially increased albedo) and consequences of nuclear war.

2. Potential direct impact and feedback processes relevant in meteorology chemistry coupling

Direct impacts of meteorology on chemistry or vice versa as well as feedback processes are varied. Their calculation only became possible only with the introduction of online meteorology chemistry models. Traditionally, aerosol feedbacks have been neglected in Met and AQ models mostly due to a historical separation between these communities, as well as a limited understanding of the underlying interaction mechanisms and associated complexities. Such mechanisms may, however, be important on a wide range of temporal and spatial scales (hours to decades and local to global). Field experiments and satellite measurements have shown that chemistry dynamics feedbacks exist among the Earth system components including the atmosphere (e.g. Kaufman and Fraser, 1997; Rosenfeld, 1999; Rosenfeld and Woodley, 1999; Givati and Rosenfeld, 2004; Jacobson, 2005; Lau and Kim, 2006; Rosenfeld et al., 2007, 2008).

The potential impacts of aerosol feedbacks can be broadly explained in terms of four types of effects: direct, semi-direct, first indirect and second indirect. For example, the reduction in solar radiation reaching the Earth by aerosols is an example of direct effect (Jacobson et al., 2007). Changes in surface temperature, wind speed, relative humidity, clouds and atmospheric stability that are caused by absorbing aerosols are examples of the semi-direct effect (Hansen et al., 1997). A decrease in cloud drop size and an increase in cloud drop number as a result of aerosols in the atmosphere are named first indirect effect (Twomey, 1977). These changes might enhance cloud albedo. An increase in liquid water content, cloud cover and lifetime of low level clouds and suppression or enhancement of precipitation are examples of the second indirect effect (Albrecht, 1989). However, this simplified classification is insufficient to describe the full range of two-way, chains and loops of interactions between meteorological and chemical processes in the atmosphere. It should also be noted that these definitions are not always consistently used throughout the literature.

The main meteorology and chemistry/aerosol interacting processes and effects, which could be considered in online coupled MetM-CTMs, are summarised in Tables 1 and 2. The order in Table 1 does not reflect their importance or relevance, since their actual relevance depends on the model application. In addition to looking at the meteorological parameters affecting chemistry, it is also worth mentioning effects of altered meteorology on meteorology, in order to better understand chains and loops of interactions. For example, clouds modulate boundary layer outflow/inflow by changes in the radiative fluxes as well as alterations of vertical mixing and the water vapour modulates radiation. The temperature gradient influences cloud formation and controls turbulence intensity and the evolution of the atmospheric boundary layer (ABL). Similar feedback mechanisms exist for altered chemistry impacts on chemistry. For

example, biogenic emissions affect the concentrations of ozone and secondary organic aerosols. The polymerisation of organic aerosols produces long chain secondary organic aerosol (SOA) with lower volatility.

Table 1. Meteorology's impacts on chemistry.

Temperature	Modulates chemical reaction and photolytic rates
	Modulates biogenic emissions (isoprene, terpenes, dimethyl sulfide, etc.)
	Influences biogenic emissions (isoprene, monoterpenes)
	Influences the volatility of chemical species
	Determines aerosol dynamics (coagulation, condensation, nucleation)
Temperature vertical gradients	Determines vertical diffusion intensity
Temperature & humidity	Affect aerosol thermodynamics (e.g., gas-particle partitioning, secondary aerosol formation)
Water vapour	Modulates OH radicals, size of <i>hydrophilic aerosol</i>
Liquid water	Determines wet scavenging and atmospheric composition
Cloud processes	Affects mixing, transformation and scavenging of chemical compounds
Precipitation	Determines the wet removal of trace gases and aerosol
Land surface parameterization (soil type and vegetation cover, soil moisture, leaf area)	Affects natural emissions (e.g. dust, BVOCs) and dry deposition
Lightning	Determines free troposphere NO _x emissions
Radiation	Determines photolysis rates and influences many chemical reaction rates
	Determines isoprene emissions
Wind speed and direction	Determines horizontal transport and vertical mixing of chemical species
	Influences dust and sea-salt emissions
ABL height	Influences concentrations

Table 2. Chemical species' impacts on meteorology.

Aerosols	Modulate radiation transfers (SW scattering/absorption, LW absorption, LW scattering by large particles like dust)
	Affect boundary layer meteorology (temperature, humidity, wind speed and direction, ABL height, stability)
	Extraordinary high concentrations can affect stability and wind speed
	Influence cloud formation, since they act as cloud condensation nuclei
Aerosols physical properties (size distribution, mass and number concentrations, hygroscopicity)	Influence cloud droplet and crystal number and hence cloud optical depth and hence radiation
	Modulate cloud morphology (e.g. reflectance)
	Influence precipitation (initiation, intensity)
	Affect haze formation and atmospheric humidity
	Influence scattering /absorption
Soot deposited on ice	Influences albedo
Radiatively active gases	Modulate radiation transfers

On a more general level, a number of chains and loops of interactions take place and should be properly simulated in an online coupled model. These may include: (a) A loop feedback starting with temperature that affects chemistry and thus chemical concentrations (Table 1); the changes in chemical concentrations will in turn affect radiative processes (Table 2), which will then affect temperature to close the loop. (b) A chain feedback starting with aerosol that affects radiation (Table 2) and thus photolysis and chemistry (Table 1). (c) A chain feedback starting with temperature gradients that affects turbulence mixing (Met-Met feedback); thus affecting surface-level pollutant concentrations (Table 1) and boundary layer outflow/inflow (Met-Met feedback). (d) A chain feedback starting with aerosols that affect cloud optical depth through influence of droplet number on mean droplet size (Table 2); the resulting changes in cloud formation will then affect the initiation of precipitation (Met-Met feedback). (e) A chain feedback starting with aerosol absorption of sunlight which results in changes in the temperature profile of the atmosphere and vertical mixing (Table 2) and thereby changes in the cloud droplet formation, which affects cloud liquid water and thus cloud optical depth (Met-Met).

Against the backdrop of the separate development of MetMs and CTMs together with the continued increase in computing power, a more detailed modelling description of physical and chemical processes and their interactions calls for a strategic vision. Such a vision will help to provide shared goals and directions for the research and operational communities in this field, while still having a multiple model approach to respond to diverse national and world-wide mandates.

3. Major challenges and needs

3.1 Interacting processes and feedback mechanisms

The focus on integrated systems is needed, since recent research has shown that interactions between meteorology and chemistry and feedback mechanisms are important in the context of many research areas and applications, which can broadly be separated into the fields of NWP, air quality/CWF and climate/earth system modelling. The relative importance of online integration and the level of detail necessary for representing different processes and feedbacks will vary greatly between the three mentioned application fields, as was also confirmed in an expert poll conducted among the members of the EuMetChem COST Action (Baklanov et al., 2014; Kong et al., 2014).

The processes which are particularly critical for online coupling between the chemical and meteorological components include (i) *advection, convection* and *vertical diffusion* (which control the transport and dispersion of chemical species and hence critically affect surface concentrations); (ii) *cloud microphysics* (which determines cloud life cycle, interactions between clouds and aerosols and affects soluble chemical species); (iii) *radiative transfer* (which is determined by meteorological parameters and radiatively active chemical compounds); and (iv) *turbulent fluxes at the surface* (which influences transport and distribution of chemical species).

Convection and condensation schemes need to be updated to take the aerosol–microphysical interactions into account, and the radiation scheme needs to be modified to include the aerosol effects more accurately. The interactions of aerosols with gas phase chemistry and their impacts on radiation and cloud microphysics depend strongly on their physical and chemical properties. Several processes such as nucleation, coagulation,

condensation, evaporation, sedimentation, in-cloud and below-cloud scavenging, and deposition at the surface need to be taken into account by the models. The aerosol–cloud interaction schemes used in models are still very uncertain, sometimes giving substantially different forcing and thus need to be improved and further developed (for example, for ice forming nuclei, interaction with cirrus clouds, contribution of different anthropogenic and biogenic/natural aerosol particles for cloud evolution). On the other hand, the inclusion of aerosol effects in convective parametrisations is only beginning to receive attention.

Online coupling imposes additional requirements on the setup and implementation of radiation parametrisations. Most of these requirements reflect the need to maintain physical and numerical consistencies between the various modules and computational schemes of the model, against the increased frequency of interactions and the multitude of simulated effects. The complexity of the treatment of the effect of simulated aerosol concentrations on shortwave and longwave radiation fluxes differs strongly among the models. A final recommendation on how complex the parametrisation needs to be is currently not possible.

Finally, emissions and deposition also interact in a specific way with the meteorological part within online coupled models. The most interesting emissions are those which depend on meteorology as they could potentially be treated more accurately and consistently than in offline models. Natural emissions (e.g. isoprene, terpenes and pollen) strongly depend on meteorology and are in general already calculated online even in offline models using the meteorological input driving the CTM model. Sea spray is the dominant aerosol source over the oceans and therefore, its proper quantification is highly relevant for a coupled model. Wind-blown dust refers to particles from a broad range of sources. Due to their direct relationship with meteorology, such emissions must be calculated online.

A large variety of chemical mechanisms are currently in use in online coupled models. Nevertheless, the most commonly-used mechanisms have converged in terms of the state of the science included in their formulation. Modifications of the chemical mechanisms, which not only affect gas phase chemistry but also the coupling with aqueous-phase and aerosol mechanisms, have faced practical difficulties in the past, requiring significant reprogramming. Methods of updating chemical mechanisms make updates much easier as illustrated in the MECCA module (Sander et al., 2005). Therefore, the following actions are recommended:

- Create a unified central database of chemical mechanisms, where mechanism owners can upload relevant codes and provide updates as necessary. Versions should be numbered and chemical mechanisms should be open.
- Enable interfacing of this database using, e.g. the Kinetic Pre-Processor (KPP) to develop a set of box model intercomparisons including evaluation against smog chamber data and more comprehensive mechanisms and moreover an analysis of the computational cost.

3.2 Numerical and computational aspects

The most relevant properties to be considered when developing integrated models and especially for considering feedback mechanisms are conservation, shape-preservation and prevention of numerical mixing or unmixing. Traditionally, Eulerian flux-based schemes are more suitable for mass conservation. Recently however, several semi-Lagrangian schemes have been developed that are inherently mass conservative. Such schemes are applied in some integrated models.

A detailed analysis of the numerical properties of European integrated models is recommended. A particularly relevant set of tests has been described by Lauritzen and Thuburn (2011), which shifts the focus from traditional, but still important, criteria such as mass-conservation to the prevention of numerical mixing and unmixing. Not maintaining the correlations between transported species is similar to introducing artificial chemical reactions in the system.

A clear trend towards integrated model development is becoming perceptible with several modelling systems that can be considered as online integrated models with main relevant feedbacks implemented. Complementing those, there are several ones that are built using an online integrated approach, but some major feedbacks are not included yet. A third group of models, the online access models, is characterised by applying an external coupler between meteorology and chemistry. All the information is passed through the coupler. Depending on the approach used, wind and mass consistency problems may arise. In this sense, online integrated models are desirable for a better representation of feedback processes.

Numerical performance is also an important issue for online models. The current parallelisation is based on well-established MPI and OpenMP programming models. Beyond these approaches there is no clear trend towards new parallelisation paradigms, even though supercomputers are experiencing a huge increase in computing power achieved mainly through an increase in the number of computing units rather than an increase in clock frequency. New processor types such as GPU's and MIC's are only beginning to be explored.

To adopt newer technologies, a conversion program that transfers existing code to the new technology would be advantageous. The transferred code would need to be still readable and maintainable. This would be very useful since a coupled meteorology chemistry model takes several decades of work to develop, and without software based support, transfers can take years to be completed reliably.

3.3 Data assimilation

Experience with chemical data assimilation (CDA) in integrated online models is still limited (see an overview in Seigneur et al., 2014). Most applications of CDA use CTMs, rather than online coupled models, to improve the simulated concentration fields or model parameters such as emissions. First efforts have been made with integrated systems (IFS-MOZART and WRF-Chem) to assimilate chemical and meteorological observations in online integrated models. There is some evidence that CDA can also improve the assimilated meteorological variables, for example the assimilation of ozone can have a positive effect on the assimilated wind fields (Semane et al, 2009). CDA will be beneficial in online coupled model if it improves the realism of the chemical fields which are used to simulate the interaction between atmospheric composition and meteorology. The easiest approach is the adjustment of initial conditions through CDA in a manner similar to meteorological data assimilation. Optimal interpolation, variational approaches, EnKF or hybrid techniques combining the advantages of both variational and EnKF techniques are applicable. Other methodologies such as inverse modelling of emission fields appear as a promising technique to improve the skill of online integrated models and may have a stronger impact for short-lived pollutants than CDA has on initial conditions. However, it is debatable whether the results of inverse modelling should be used directly to correct emission fields or only to provide insights for the development of improved emission inventories.

3.4 Evaluation of methodologies and data

There is a crucial need for more advanced evaluation of methodologies and output data. Model validation and benchmarking are important elements of model development as they help identifying model strengths and weaknesses. Model validation has a long tradition in the NWP and AQ modelling communities, and many concepts can be applied to online integrated models as well. The MetM community has the necessary tools, for example, to analyse whether including certain feedbacks or not has a positive effect on weather forecast skill. Demonstrating these benefits however, requires running a model with and without feedbacks over extended periods of time - rather than for selected episodes - in order to draw statistically significant conclusions.

Evaluating whether relevant feedback processes are treated accurately by a model is challenging. The effects of aerosols on radiation and clouds, for example, depend on the physical and chemical properties of the aerosols. Thus, comprehensive measurements of aerosol size distributions, chemical composition, and optical properties are needed. Such observations should ideally be collocated with detailed radiation measurements (e.g. WMO GAW, AERONET), with aerosol lidars probing the vertical distribution and with radiosondes providing profiles of temperature and humidity. Evaluating indirect aerosol effects on clouds and precipitation is even more challenging and requires additional detailed observations of cloud properties such as cloud droplet number concentrations. Measurements from polarimetric radars, disdrometers, and cloud particle imagers can provide information on hydrometeor phases and size distributions but are only sparsely available. Online integration can also be beneficial for AQ modelling. Dense observational networks are available for the validation of classical air pollutants such as O_3 or NO_x and satellite observations of AOD and NO_2 .

4. Future directions, perspectives and recommendations

It is clear that the online modelling approach is a prospective way for future *single-atmosphere* modelling systems, providing advantages for all three communities, Met modelling including NWP, AQ modelling including CWF, and climate modelling. However, there is not necessarily one integrated online modelling approach/system suitable for all communities.

Comprehensive online modelling systems, built for research purposes and including all important mechanisms of interactions, will help to understand the importance of different processes and interactions and to create specific model configurations that are tailored for their respective purposes.

Regarding **CWF and AQ modelling** the online approach will certainly improve forecast capabilities as it allows a correct way of jointly and consistently describing meteorological and chemical processes within the same model timesteps and grid cells. This also includes harmonised parametrisations of physical and chemical processes in the ABL. There are many studies and measurements supportive of this conclusion (Grell et al., 2004; Grell, 2008; Zhang, 2008; Korsholm et al., 2009; Grell and Baklanov, 2011; Forkel et al., 2012; Saide et al., 2012; Zhang et al., 2013). In particular, due to the strong nonlinearities involved, offline coupling can lead to inaccuracies in chemical composition simulations.

For **meteorological modelling**, the advantages of online approaches are less evident and need to be further investigated and justified. It is clear that online models for short-term applications like NWP do not require full comprehensive chemistry (which would increase the CPU cost tremendously). Rather, the main improvements for NWP that are possible through an online integrated approach will be related to improvements in (i) meteorological data assimilation (first of all remote sensing data, radiation characteristics, which require detailed distributions of aerosols in the atmosphere) and (ii) description of aerosol–cloud and aerosol–radiation interactions, yielding improved forecasting of precipitation, visibility, fog and extreme weather events. While these improvements might not be statistically significant as averaged over longer periods of time, it is clear that for specific episodes and for urban weather forecasts, there are large potential benefits. In summary, meteorology modelling including NWP should benefit from including such feedbacks as aerosol–cloud–radiation interactions, aerosol dynamics along with simplified chemistry (with a focus on aerosol precursors and formation, e.g. sulfur chemistry).

For **climate modelling**, the feedbacks (forcing mechanisms) are the most important and the main improvements are related to climate–chemistry/aerosols interactions. However, the online approach is not strictly necessary for all purposes in this field. Many GCMs or RCMs are using an offline approach for describing GHG and aerosol forcing processes (by chemistry/aerosol parametrisations or prescription or reading outputs of CTMs). For global climate, in the EU project MEGAPOLI, a sensitivity study compared online vs. offline approaches and showed that for long-lived GHG forcing the online approach did not give large improvements (Folberth et al., 2011). On the other hand, for short-lived climate forcers, especially aerosols and for regional or urban climate, the outcome was very different, with online modelling being of substantial benefit. The online approach for climate modelling is mostly important for studies of short-lived climate forcers, which represent one of the main uncertainties in current climate models and are in particular at the core of political and socio-economic assessments of future climate change mitigation strategies. It will be impossible to answer the main questions about aerosol short-lived climate forcers and mitigation strategies without employing fully online coupled modelling systems that include aerosol dynamics and feedbacks.

Based on several overview analysis of the models (Zhang, 2008; Grell and Baklanov, 2011; Kukkonen et al., 2012; Zhang et al., 2012a,b; Baklanov et al., 2014), we suggest aiming at eventually migrating from separate MetM and CTM systems to online integrated coupled meteorology chemistry models. Only this type of model allows the consideration of two-way interactions (i.e., feedbacks) in a consistent way. The integration has not only the advantage of a single-atmosphere model, for instance where water vapour and other atmospheric gases are no longer treated numerically differently simply because of historical separation of the different disciplines. Furthermore, the integration has the advantage of saving computational resources, since several processes (e.g. vertical diffusion) have to be described in both MetMs and CTMs. Moreover, it will also reduce the overall efforts in research and development, maintenance and application leading to cost savings for both types of models.

The main recommendations are briefly summarised in the following sections. If a recommendation is mainly relevant for one type of the application (Meteorology or Chemistry simulations), this is explicitly mentioned.

4.1 Emissions and depositions

Emission and deposition are both close-to-surface processes and dependent on meteorology processes. In order to improve their treatment in MetChem models the following is needed:

- Time dependence of anthropogenic emissions should be better described, and open-ocean ship emissions should be better characterised (time, amount, compounds). Currently their parametrisations still have large uncertainties (Jalkanen et al., 2012).
- Accurate characterisation of land use, soil moisture and vegetation should be used for more accurate representations of meteorologically-dependent emissions.
- Emissions and heat fluxes from forest fires and volcanic eruptions need to be better known and improved in the models.
- Treatment of anthropogenic VOC emissions need to be improved/updated, both because of their contributions to O₃ and SOA formation.
- Emissions of primary aerosols and in particular their number and size distributions and physical properties (hydrophilic/hydrophobic) need to be better represented, both for atmospheric composition and for interaction with meteorology.
- Ammonia emissions should be calculated online with a more accurate representation of temporal variation, and account should be taken of their interactions with soil/vegetation (bi-directional fluxes, deposition or emission).
- Dry and wet deposition processes are directly driven by meteorology and, therefore, more accurate representations of their interplays with chemistry and meteorology are needed.
- Accurate parametrisations of land surface processes and accurate land use/land cover data sets are needed because of their profound impacts on both natural emissions and dry deposition fluxes.

4.2 Model formulations

Migrate from offline to online integrated modelling systems is recommended as only the latter approach can guarantee a consistent treatment of processes and allow two-way interactions of physical and chemical components of Met-Chem systems, particularly for CWF and NWP communities. Online integrated models, however, need harmonised formulations of all processes influencing meteorology and chemistry. In particular the following model treatments need to be considered:

- Our parametrisation/understanding of aerosol–radiation–cloud–chemistry interactions is still incomplete and further research on the model representations of these interactions is needed.
- Key aerosol properties (size distribution, phase, hygroscopicity, mixing state and optical depths) and processes (chemistry, thermodynamics for SOA and dynamics) need to be better represented for AQ simulations.
- Cloud properties (droplet number concentrations, size distribution, optical depths), processes (microphysics, dynamics, wet scavenging, aqueous phase chemistry) and cloud–aerosol interactions for all types of clouds (in particular for convective and ice clouds) need to be better represented.
- A major challenge for most online models is the adequate treatment of indirect aerosol effects. Its implementation with affordable computational requirements and evaluation against laboratory/field data would greatly facilitate this transition.

- As more meteorological and chemical variables are assimilated into a model, one must be cautious about possible diminishing returns and possible antagonistic effects due to the interactions between meteorological variables and chemical concentrations. Consequently, the development of optimal methods for data assimilation is warranted.

4.3 Real-time application

To achieve the objective of online coupled meteorology and chemistry simulation in forecast models some specific aspects should be considered:

- National weather centres should consider progressively including aerosol-chemistry interactions into NWP systems which will lead to potential improvements and extending them to CWF using online coupled models for cross evaluations, benefitting both disciplines.
- The online integrated approach is well suited for applications where a frequent integration between meteorology and chemistry models is required to properly account for the effects of mesoscale events in high-resolution CTMs.
- The online coupling of meteorology, physics and emissions and their accurate representations are essential for CWF; the implementation of aerosol feedbacks is important mostly for specific episodes and extreme cases.

4.4 Model evaluation

For online models the evaluation can no longer be conducted for meteorology or chemistry separately. Interacting processes will need specific attention to avoid the situation where the “right” results are obtained for the wrong reasons. In this regard, efforts should focus on conducting dynamic evaluation to establish the models’ credibility in accurately simulating the changes in weather and air quality conditions observed in the real world. To achieve this, attention should be given to:

- An international test bed for evaluation of urban models and mesoscale models for online MetChem models. A first step into this direction has been taken by the AQMEII consortium for the regional scale, but extension for higher resolving models is important.
- Special variables (e.g. shortwave and longwave radiation, photolytic rate of NO₂, AOD, COT, CCN, CDNC, precipitation) should be included routinely into a model evaluation for online coupled models. Reliable measurements are needed on a routine basis.
- Routine, long-term measurements of aerosol size distributions, chemical composition and optical properties in operational ground-based networks are urgently needed to verify meteorology/climate–chemistry feedbacks.
- Ground-based and satellite remote-sensing measurements of aerosol and cloud properties (e.g. optical depths, CCN, IN, CDNC and SW and LW radiation) are very important to study aerosol indirect effects and should be included for validation of meteorology chemistry feedbacks.
- Last but not least, there is a need to evaluate routinely the atmospheric mixing processes in models, in particular within the ABL, using measurements on fluxes of meteorological parameters and chemical species in all three directions.

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