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Online coupled regional meteorology-chemistry models in Europe: current status and prospects

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Abstract

The simulation of the coupled evolution of atmospheric dynamics, pollutant transport, chemical reactions and atmospheric composition is one of the most challenging tasks in environmental modelling, climate change studies, and weather forecasting for the next decades as they all involve strongly integrated processes. Weather strongly influences air quality (AQ) and atmospheric transport of hazardous materials, while atmospheric composition can influence both weather and climate by directly modifying the atmospheric radiation budget or indirectly affecting cloud formation. Until recently, however, due to 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.

The continuous increase in computer power has now reached a stage that enables us to perform online coupling of regional meteorological models with atmospheric chemical transport models. 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 numerical weather prediction (NWP), AQ forecasting as well as climate and Earth system modelling. However, the relative importance of online integration and its priorities, requirements and levels of detail necessary for representing different processes and feedbacks can greatly vary for these related communities: (i) NWP, (ii) AQ forecasting and assessments, (iii) climate and earth system modelling. Additional applications are likely to benefit from online modelling, e.g.: simulation of volcanic ash or forest fire plumes, pollen warnings, dust storms, oil/gas fires, geo-engineering tests involving changes in the radiation balance.

The COST Action ES1004 – European framework for online integrated air quality and meteorology modelling (EuMetChem) – aims at paving the way towards a new generation of online integrated atmospheric chemical transport and meteorology modelling with two-way interactions between different atmospheric processes including dynam-

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ics, chemistry, clouds, radiation, boundary layer and emissions. As its first task, we summarise the current status of European modelling practices and experience with online coupled modelling of meteorology with atmospheric chemistry including feedback mechanisms and attempt reviewing the various issues connected to the different modules of such online coupled models but also providing recommendations for coping with them for the benefit of the modelling community at large.

1 Introduction

The prediction and simulation of the coupled evolution of atmospheric dynamics, pollutant transport, chemical reactions and atmospheric composition will remain one of the most challenging tasks in environmental modelling, climate change studies, and weather forecasting 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 recognized that atmospheric composition can influence both weather and climate directly by changing the atmospheric radiation budget or indirectly by affecting cloud formation. 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.

The dramatic increase in computer power during the last decade is enabling us to reach high spatial resolution (say, a few km) in numerical weather prediction (NWP) and meteorological modelling. Fronts, convective systems, local wind systems, and clouds are being resolved or partly resolved. Furthermore, the complexity of the parameterisation 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 regional meteorological models (MetM) with atmospheric chemical transport models (CTM) either offline or online (Fig. 1). Offline modelling implies that the CTM is run after

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the meteorological simulation is completed, while online modelling allows coupling and integration of some of the physical and chemical components to various degrees. The ultimate stage is to fully couple the CTM with the MetM to produce a unified modelling system with consistent treatment of processes such as advection, turbulence, and radiation for both meteorological and chemical quantities. This allows generating online fully integrated meteorology-chemistry simulations (OCMC) with two-way interactions (i.e., feedbacks).

Climate modelling is also expanding its capacities through the use of an earth system modelling approach that integrates atmospheric chemistry, the oceans and the biosphere. Climate modelling, however, does not require the implementation of near-real-time data assimilation, which is crucial for the skill of NWP and can help improve AQ forecasts.

However, combining two systems which both have high CPU and memory requirements for operational applications still poses many problems in practice and, thus, is not always feasible at NWP 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 weather 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 OCMC approach is already used in many research-type atmospheric models.

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

Historically, Europe has not adopted a community approach to modelling and this has led to a large number of model development programmes, usually working al-

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most independently, thereby yielding results tailored for specific applications. However, a strategic framework could help provide a common goal and direction to European research in this field, while still having various models as part of an ensemble. The task is many-fold since it requires scientific knowledge and practical experience in NWP, AQ modeling, numerics, atmospheric physics and 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, 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 detail necessary for representing different processes can greatly vary between applications and therefore tailored solutions may be required for the three communities: (i) NWP, (ii) AQ forecasting and assessments, (iii) climate and earth system modelling.

For example, NWP does not require detailed chemical processes, but aerosols, via radiative and microphysical processes can affect fog formation, visibility and precipitation and hence 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 definitely improves AQ and chemical atmospheric composition projections. These modelling communities have different targets with respect to temporal as well as spatial scales, but also as to the processes under focus. For AQ forecasting, the key issue is usually the ground-level concentration of pollutants, whereas for weather and climate models, skill is typically based on screen level temperature, precipitation and wind.

Several applications are likely to benefit from online modelling though they do not clearly belong to one of these three main communities, e.g.: bio-weather forecasting, pollen warnings, forecasting of hazardous plumes from volcanic eruptions, forest fires and oil and gas fires, dust storms, assessing geo-engineering techniques that involve

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changes in the radiation balance (e.g. input of sulphate aerosols, artificially increased albedo), nuclear war consequences, etc.

This paper summarises the current status of modelling practices towards online coupled modelling of meteorology with atmospheric chemistry with a specific focus on European models and research. Section 2 is a survey of the potentially relevant processes in the interactions between atmospheric dynamics (meteorology) and atmospheric composition (AQ, climate). Section 3 gives a brief overview of European developments and existing online mesoscale models. Section 4 describes how feedback processes are treated in these models. Section 5 addresses the numerical issues of coupled models. Section 6 describes a few case studies and model evaluation methods. Conclusions are finally drawn in Sect. 7. Appendix A includes brief descriptions of the main regional online coupled or integrated models, developed or actively used in Europe. A list of acronyms is also given at the end.

This paper focuses on models resolving mesoscale phenomena, thus with grid-sizes between 1 km and about 20 km, but does not cover global or local models. Furthermore, the time scale of interest is for simulations of episodes (1-day to a week) rather than for climate simulations. Therefore, some aspects rated here as less relevant might be of much more importance for climate models (e.g., changes in biodiversity due to nutrient load with impacts on evaporation, surface albedo, etc.).

2 Survey of potential direct and feedback processes relevant in meteorology-chemistry coupling

Traditionally, aerosol feedbacks have been neglected in MetM 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, from days to decades and from local to global. Field experiments and satellite measurements have shown that chemistry-dynamics feedbacks exist within the

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Earth system components including the atmosphere (e.g., Kaufman and Fraser, 1997; Rosenfeld, 1999; Rosenfeld and Woodley, 1999; Givati and Rosenfeld, 2004; Lau and Kim, 2006; Rosenfeld et al., 2007, 2008).

The potential impacts of aerosol feedbacks can be broadly segregated into four types (Jacobson et al., 2007; Baklanov et al., 2007; Zhang, 2008; Zhang et al., 2010a, 2012c; Grell and Baklanov, 2011):

- A reduction in solar radiation reaching the Earth (direct effect);
- Changes in surface temperature, wind speed, relative humidity, and atmospheric stability (semi-direct effect);
- A decrease in cloud drop size and an increase in cloud drop number which enhances cloud albedo (first indirect effect);
- An increase in liquid water content, cloud cover, and lifetime of low level clouds, and suppression or enhancement of precipitation (second indirect effect).

However, this simplified classification is insufficient to describe the full range of two-way and loop interactions between meteorological and chemical processes in the atmosphere. 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.

Additionally it is worth mentioning a number of mechanisms of altered meteorology impacts on meteorology, e.g.:

Clouds → Modulate boundary layer outflow/inflow,

Water vapour → Modulates radiation,

Temperature gradient → Influences cloud formation and controls turbulence intensity;

and of altered chemistry impacts on chemistry, e.g.:

Biogenic emissions → Affect concentrations of ozone and secondary organic aerosols,

Polymerisation of organic aerosols → Yields long chain SOAs with lower volatility.

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On a more general level, the following chains of interactions take place and should be properly simulated in an OCMC model:

- Temperature → chemistry → concentrations → radiative processes → temperature (Fig. 2);
- Aerosol → radiation → photolysis → chemistry;
- Temperature gradients → turbulence → surface concentrations, boundary layer outflow/inflow;
- Aerosol → cloud optical depth through influence of droplet number on mean droplet size → initiation of precipitation;
- Aerosol absorption of sunlight → cloud liquid water → cloud optical depth.

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

With this perspective, 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 and meteorology modelling. The Action is not aimed at determining or designing one best model, but to identify and review the main processes and to specify optimal modular structures for integrated Meteorology-Chemistry (MetChem) models to simulate specific phenomena. Furthermore, the COST Action will develop recommendations for efficient interfacing and integration of new modules, keeping in mind that there is no one best model, but that an ensemble of models is likely to provide the most skillful forecasts.

One of the initiatives of this COST Action was to perform an expert poll to identify the most important chemistry–meteorology interactions (as listed in Tables 1 and 2) and

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how they are represented in current models. The survey design was similar to the expert poll in the EU FP7 PEGASOS project (http://pegasos.iceht.forth.gr/) but extended to cover three model categories: NWP, CWF and climate models. This survey not only rates the importance of the meteorology-chemistry interactions, but also rates how well they are represented in current online models. The survey questionnaire was sent to different experts in these communities in Europe and beyond, and the results of its analysis (based on 30 responses) are shown in Table 3.

These results show that the perceived most important interactions differ from one model category to another. In general, most of the meteorology and chemistry interactions are more important for CWF models than NWP and climate models, and those interactions are represented better in CWF models than in NWP and climate models (see averaged scores in Table 3). However, only a few interactions are felt to be "quite well" or "fairly well" represented in models.

Therefore, primary attention needs to be given to interactions with high rank of importance (score1) together with low score in the model representation (score2), such as "improvement of aerosol indirect effects" for both NWP and climate models, "changes in liquid water affect wet scavenging and atmospheric composition" and "improvement of wind speed – dust/sea-salt interactions" for CWF models (see highlighted rows in Table 3). However, the complexity of these interactions might hamper their improved representation directly through only one simple change. The survey results might be also affected by individual opinions. Most people have key expertise in only one or two of the models/model categories, which caused difficulties in answering some of the questions. A large percentage of "don't know" answers given for the representation of interactions in climate models is not surprising, as COST ES1004 is primarily aiming at the AQ and CWF community and there are very few climate modellers in this action.

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3 Overview of currently applied mesoscale online coupled meteorology and air quality models

L. F. Richardson (1922), the pioneer of weather prediction modelling, suggested an online coupled meteorology-pollution model by including a dust transport equation into his NWP model formulation; this was, however, not completely realised at that time. More than half a century later, online coupling started to be considered more relevant, e.g., at the Novosibirsk scientific school in the USSR (Marchuk, 1982; Penenko and Aloyan, 1985; Baklanov, 1988) or in the German non-hydrostatic modelling community (GESIMA, Eppel et al., 1995; MESOSCOP, Alheit and Hauf, 1992; METRAS, Schlünzen and Pahl, 1992; see Schlünzen, 1994, for an overview of that period). The earliest online approach for the simulation of climate, air quality and chemical composition may have been a model developed by Jacobson (1994, 1996). However, the online calculation was still very expensive and, at most, only online access models were used (e.g., aerosol composition change studied for a coastal area by von Salzen and Schlünzen, 1999c). Online integration with inclusion of a wider range of feedbacks has become possible only in the last decade.

The main characteristics of online and offline approaches are discussed in different papers, e.g.: Peters et al. (1995), Zhang (2008), Grell and Baklanov et al. (2011). We will proceed from the following definitions of offline and online models (Baklanov et al., 2007):

3.1 Offline models

- Separate CTMs driven by meteorological input data from meteo-preprocessors, measurements or diagnostic models,
- Separate CTMs driven by analysed or forecasted meteorological data from MetM archives,

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– Separate CTMs reading output-files from operational NWP models or specific MetMs with a limited period of time (e.g., 1, 3, 6 h).

3.2 Online models

– Online access models, in which meteorological and chemical data are available at each time-step but CTM and MetM are separate models linked via a model interface,

– Online integration of the CTM into a MetM, when the CTM is called each time-step inside the MetM and processes are treated consistently for both chemical and meteorological quantities.

Feedbacks between meteorology and chemistry are not possible in offline models but are typically included in online integrated models and sometimes in online access models.

American, Canadian and Japanese institutions have developed and used online coupled models operationally for AQ forecasting and for research (GATOR-MMTD: Jacobson et al., 1996, 1997a,b; WRF-Chem: Grell et al., 2005; GEM-AQ: Kaminski et al., 2007; WRF-CMAQ: Mathur et al., 2008). The European Centre for Medium Range Weather Forecasting (ECMWF) also noted the relevance of online chemistry (Hollingsworth et al., 2008) and several national weather services (DMI, UK MetOffice, etc.) and research institutions (KIT, ISAC, etc.) are developing online models. For operational or climate applications the cost of online integration is still an issue. Therefore it is important to determine which feedbacks have the largest impact, and what are the minimum requirements to accurately represent them in an online integration.

The atmospheric model database initiated within the previous COST Action 728 (http://www.mi.uni-hamburg.de/costmodinv) and the related overview by WMO and COST-728 (Baklanov et al., 2007, 2011a; Schlünzen and Sokhi, 2008) show a number of online coupled MetM and CTM systems being developed and used in Europe. Great progress has been made during the past 5 yr with currently more than 20 online cou-

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pled modelling systems in use; in 2007 – only one European model considered aerosol indirect feedbacks (Enviro-HIRLAM), now – about 10. The list and current status of online access or online integrated MetChem models developed or applied in Europe are presented in Table 4. These models use grid-sizes ranging from 1 to 20 km. We define here the length of an episode as ranging between 1 day to 1 week, while the long-term horizon addresses integrations over periods of more than 1 week. Short descriptions of these models are given in Appendix A with some examples of their main applications. Further assessment of online MetChem models in the subsequent sections of this paper will involve mostly these listed models as examples.

4 Current treatments of interacting processes in online-coupled models

4.1 Meteorological model cores: dynamical and physical processes – interaction with chemistry

The wide range of coupled chemistry–meteorology models used in Europe is based upon an equally large variety of meteorological model cores. The basic theoretical descriptions of meteorological processes are widely described in meteorology textbooks and, hence, beyond the scope of this paper. This section reviews the processes typically used in current NWP models, to which these coupled models are linked.

The set of equations which are solved in these models fall broadly into two categories: (1) the *dynamics* core that calculates the evolution of the atmospheric flow via grid-resolved processes; and (2) the *physics* core that usually includes the unresolved fluid dynamical processes (e.g., boundary layer turbulence, subgrid-scale orographic drag, non-orographic gravity wave drag, convection) and non-fluid dynamical processes (e.g., radiation, clouds and large-scale precipitation, surface-atmosphere interactions). NWP models differ greatly in terms of their treatment of dynamical and physical processes, their discretization schemes, approximations (hydrostatic vs. non-hydrostatic) as well as their advection formulation (semi-Lagrangian vs. Eulerian).

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The dynamical and physical processes that are relevant for coupling meteorology and atmospheric chemistry (i.e., which have a strong *direct* influence on atmospheric composition) include:

1. *Advection* is a grid-resolved process in meteorological models, which largely controls the atmospheric transport of chemical species in coupled models. Mass conservation can become an issue if meteorological and chemical variables are not advected using the same numerics. There are large differences among models, with Eulerian and semi-Lagrangian schemes as the two main classes (see Sect. 5.1). Eulerian schemes can be made trivially conservative (e.g., weighted average flux methods; Toro, 1992), but sophisticated methods have been developed also for semi-Lagrangian formulations (e.g., Kaas, 2008).

2. *Convection* is a parameterized sub-grid process affecting surface concentrations and vertical distribution of chemical species in coupled models. It is often divided into shallow, mid-level and deep convection. Several numerical schemes are widely used (Tiedtke, 1989; Kain and Fritsch, 1993; Zhang and McFarlane, 1995; Manabe et al., 1965; Grell and Devenyi, 2002).

3. *Vertical diffusion* is typically implemented through solving the advection-diffusion equation using diffusion coefficients computed with different methods. Some are diagnostic, while others are based on prognostic equations for the turbulent kinetic energy (TKE) and a diagnostic estimation of the mixing length scale. One of the critical questions for the coupling is the atmospheric boundary layer (ABL) height. It is a quantity that is not always well-defined (Seibert et al., 2000), at least, in stable cases, but which has great influence on surface concentrations (Dandou et al., 2009; Schäfer et al., 2011). Stable cases are the most problematic, and large differences among models can appear (Zilitinkevich and Baklanov, 2002; Svensson et al., 2011).

4. *Cloud microphysics* determines the formation and lifetime of clouds and has important effects on chemical (water-soluble) species in coupled models. Cloud

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schemes usually also take into account important cloud processes such as cloud-top entrainment, precipitation of water and ice and evaporation of precipitation (see Sect. 4.4 and overview in Stensrud, 2007; Sokhi et al., 2013).

5. *Radiation schemes* calculate fluxes from temperature, specific humidity, liquid/ice water content and cloud fraction, and radiatively-active chemical components. These should include black and organic carbon (BC, OC), sulphate, sea-salt, dust, and other aerosols as well as the main greenhouse gases (GHG) such as CO_2, O_3, CH_4, N_2O, $CFCl_3$ and CF_2Cl_2. Major issues are how accurately these species are represented and how refractive indices are defined for aerosols (internal/external mixing, etc.) (e.g., overview in Stensrud, 2007; Sokhi et al., 2013).

6. *Drag* interactions with the surface are parameterised through different surface layer formulations (e.g. Louis, 1979; Zilitinkevich et al., 2006) and canopy (vegetation or urban) models (Hidalgo et al., 2008). Above the sea surface drag is often parameterised using the Charnock (1955) formula due to missing wave data. It works reasonably well for flat coastal regions, while for deeper water recent studies suggest a different approach (Foreman and Emeis, 2010). In particular, this latter approach has a much better asymptotic behaviour for high wind speeds and hurricanes. In some cases (e.g., ECMWF-IFS) a two-way interaction has already been established between wind and the wave model (e.g., Janssen et al., 2002).

4.2 Atmospheric chemical mechanisms: gas and aqueous phase

The chemical mechanism implemented in a model can only represent a simplified set of all the chemical reactions of the actual atmosphere. This is necessary due to the complexity of the atmospheric system for both predicting concentrations of gases or calculating the source of pollutants. The chemical reactions needed to model air quality include gas phase reactions (both thermal reactions and photo dissociation), heterogeneous reactions on surfaces (both on the surface of aerosols and other surfaces such as the ground or buildings) and aqueous phase reactions. These are coupled

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to various other processes in the atmosphere. Gas phase reactions depend on the temperature; photolysis rates depend on radiative transfer which, in turn, depends on cloud and aerosol processes; the production of the hydroxyl radical depends on the water vapour concentration. Heterogeneous reactions on aerosols depend on the surface area and composition of the aerosol and aqueous phase chemistry is a function of the cloud liquid water content. The concentrations of chemically and radiatively active gases (in particular, ozone and methane) are important for climate modelling timescales, while chemistry on aerosol formation has a greater role for meteorology on shorter timescales.

As seen in Table 6 there is a wide variety of chemistry schemes/mechanisms currently in use to simplify the chemistry to varying extent. For comparison, the current version of the Master Chemical Mechanism (MCM – Jenkin et al., 1997; Saunders et al., 2003) contains about 17 000 reactions, 6700 primary, secondary and radical species, as well as nearly 800 reactions of explicit aqueous phase mechanisms (Herrmann et al., 2005), whereas the mechanisms described operationally in 3-D models include at most a few hundred reactions (condensed or reduced mechanisms).

Since the number of species and reactions of atmospheric inorganic chemistry is manageable computationally, most of the reduction concerns the organic chemistry pathways. Although comprehensive mechanisms such as MCM have been incorporated in 3-D models (e.g., Jacobson and Ginnebaugh, 2010; Ying and Li, 2011), most 3-D models use reduced mechanisms for computational efficiency. Simplification can be achieved in a number of ways – by neglecting volatile organic compound (VOC) species of lesser importance, by removing less important chemical reactions from the mechanism or by lumping together related chemical species or functional groups. This simplification (or "mechanism reduction") can be achieved by using automated tools that analyse the mechanism (e.g., Szopa et al., 2005) or by expert knowledge. The reduced mechanism is then evaluated by comparison to laboratory "smog chamber" data and/or to a large mechanism. Most chemical mechanisms used in 3-D models have been developed independently from large mechanisms. One can distinguish two main cate-

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gories of reduced mechanisms: the surrogate molecule mechanisms and the carbon-bond mechanisms. In a surrogate molecule mechanism, several VOC molecules of the same class (e.g., short-chain alkanes) are grouped and represented by a single molecule. The mechanism associated with that molecule is then typically a weighted average of the molecules that it represents. In a carbon-bond mechanism, each VOC molecule is broken down into functional groups (e.g., carbonyl group, double-bond) and an oxidation mechanism is developed for each of those functional groups. The creation of a chemical mechanism is a significant investment. A further issue is that in different parts of the atmosphere different chemical reactions are important. In this paper, we focus on regional models and on tropospheric chemistry, but it should be noted that some global models incorporate both stratospheric and tropospheric chemistry. In general, most models use both gas and aqueous phase mechanisms, with relatively more detail in the gas phase mechanisms. The more explicit treatment of liquid phase chemistry occurs in association with limited area models that include interactions with cloud systems (Tilgner et al., 2010; Lim et al., 2010). Lim et al. (2010) show the importance of aqueous phase chemistry in the production of secondary organic aerosol (SOA).

There is a wide variety of chemical mechanisms currently in use. Among the most commonly used are the following surrogate molecule mechanisms, RADM2, RACM, RACM2, RACM-MIM, SAPRC90, SAPRC99, SPARC07TB, MELCHIOR, ADOM, MOZART2, 3 and 4, NWP-Chem, RADMK, ReLACS, RAQ, MECCA1, GEOS-Chem, and the most recent carbon-bond mechanisms, CB-IV, CB05 and CBM-Z. Some mechanisms were developed independently (e.g., SAPRC99, CB05) while others were developed in connection with a specific CTM (e.g. NWP-Chem developed for Enviro-HIRLAM, RAQ for the MetUM). Some mechanisms even carry the name of the corresponding CTM (MOZART, GEOS-CHEM). By using a mechanism developed previously, less effort is required in setting it up and in updating to account for new laboratory findings. However, the advantage of a group creating its own mechanism is that

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they can make their own judgments about the importance of specific reactions and the cost/benefit to the desired model applications.

A complication in attempting to review chemical mechanisms in use is that some mechanisms have optional sections which are included or excluded depending on the requirements of a specific study. This means that the number of tracers and reactions is not a single number for each mechanism. For mechanisms where this is true we include a range in Table 6. Furthermore, some groups take an existing mechanism and make their own modifications (for example RADMK is a modified version of RADM) or keep an existing mechanism but update the reaction rates with the latest recommended values.

Today, the most commonly-used mechanisms have converged in terms of the state of the science included in their formulation and comparable results are obtained with a surrogate molecule mechanism and a carbon-bond mechanism. Nevertheless, differences occur in the simulated concentrations, which result from differences in the oxidation mechanisms of VOC (for aromatics for example) as well as in the kinetic data (e.g., for the oxidation of NO and NO_2). Such differences have been quantified in recent studies conducted over the US (e.g., Luecken et al., 2008; Faraji et al., 2008; Zhang et al., 2012d) and Europe (Kim et al., 2009, 2011). Note that differences in gas-phase chemistry affect not only the concentrations of gaseous pollutants but also those of secondary particulate matter (PM) compounds.

Chemical mechanisms will continue to vary substantially given the different needs for different applications. However, some common issues which will likely be addressed in the near future include: correctly modelling the HO_x budget, especially in areas with high isoprene concentrations (e.g., Stone et al., 2010), the influence of aerosol composition on heterogeneous chemistry (e.g., Riedel et al., 2012), and addressing the tendency for many models to under-predict SOA (e.g., Volkamer et al., 2006; Farina et al., 2010).

During the last decade new software tools have become available, in particular the Kinetic Pre-Processors (KPP) (Sandu and Sander, 2006; Damian et al., 2002), that as-

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sist the computer simulation of chemical kinetic systems (see, e.g.: http://people.cs.vt.edu/~asandu/Software/Kpp/). They can automatically generate code for a user-defined chemical mechanism and numerical solver chosen by the user for a specific task. Tools such as KPP have the additional advantage of generating not only new mechanisms if equations or reaction rates change and new reactions are added, but also they may be able to generate adjoints. KPP makes updating of chemical mechanisms much easier as illustrated in the MECCA module (Sander et al., 2005).

As was mentioned already, the requirements and level of complexity necessary for representing different chemical processes are different for NWP, CWF and climate online modelling. For example, NWP does not depend on detailed chemical processes that may be necessary to predict air quality, but needs just enough complexity to be able to model aerosol effects on radiative and precipitation processes. A simple bulk aerosol approach may be sufficient, for example, aerosol modules from GOCART are used by WRF-Chem for NWP applications. Enviro-HIRLAM for NWP and long-term runs is developing a very simplified chemical scheme based on the ECHAM chemistry. For climate modelling, chemistry of greenhouse gases and aerosols become very important, however for long-lived GHGs, online integration of full-scale chemistry is not critically needed. For CWF and prediction of atmospheric composition in a changing climate, more advanced and comprehensive chemical mechanisms are much more important.

Future model intercomparisons would greatly benefit from the establishment of a central mechanisms database, to which mechanism owners could upload their scheme and provide further updates as necessary. This would allow true versioning and openness for chemical mechanisms. All modelling groups should be encouraged to upload their own mechanisms whenever they make changes, even if they only change the reaction rates in an existing mechanism. Ideally this could be interfaced to a set of box model inter-comparisons including evaluation against smog chamber data, field campaigns and highly complex mechanisms. It would also allow direct comparison of the computing costs of the mechanisms.

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4.3 Aerosol dynamics and thermodynamics

Aerosols differ by morphology, size, and chemical composition. They have an impact on atmospheric radiation and cloud microphysics, and they interact with gas phase chemistry. These interactions depend on size and chemical composition. In this respect, water is an important component of the aerosol particles as the water content determines the chemical composition and at the same time chemical composition determines the water content. The size range of atmospheric aerosol particles covers several orders of magnitude. Aerosols can be composed of hundreds of chemical compounds. Therefore, the numerical treatment of aerosol particles in atmospheric models needs sophisticated methods and considerable simplifications.

There are several processes modifying physical and chemical properties of aerosol particles that need to be taken into account by the models. These are nucleation, coagulation, condensation (and evaporation), sedimentation, in-cloud and below-cloud scavenging, and deposition at the surface. The approaches that are currently used in online coupled models can be classified in the following way (see Table 7).

4.3.1 Bulk approach

The simplest way to take into account aerosols in a numerical model is the so-called bulk approach, whereby aerosols are represented by mass density only. The size distribution is neglected or prescribed when necessary, and assumptions have to be made when other physical or chemical variables that depend on size or surface are treated. Still, it is possible to simulate the chemical composition of the aerosol particles. Processes like coagulation cannot be taken into account.

4.3.2 Modal approach

One possibility to simulate the size distribution is the so-called modal approach. This approach assumes, justified to some extent by observations, that real world size dis-

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tributions can be approximated by several overlapping modes, each of them described by a log-normal distribution. In principle, prognostic equations for three moments of the log-normal distributions have to be solved, e.g., for total number density, standard deviation and total mass concentration, but advection mainly hinders a consistent treatment of three moments at the same time. Therefore, in most aerosol models and for the same reason also in cloud models, prognostic equations for two moments are solved and the standard deviations of the log normal distributions are kept constant. However, this may lead to large errors (Zhang et al., 1999) and, accordingly, some models (e.g., CMAQ, Polair3D/MAM) treat the standard deviations variable to obtain better accuracy. Examples of the modal approach include: COSMO-ART with 11 modes (Vogel et al., 2009); Enviro-HIRLAM with 3 modes (Baklanov, 2003; Korsholm, 2009); MCCM, WRF-Chem with the aerosol module MADE-SORGAM (Ackermann et al., 1998; Schell et al., 2001) and MADE/VBS with 2 modes (Ahmadov et al., 2012).

4.3.3 Sectional (bin) approach

Another method to describe space and time dependent size distributions of aerosols is the so-called sectional approach. In this case, the size ranges are divided into fixed sections (or bins). As for the modal approach, processes such as nucleation, coagulation, condensation/evaporation, scavenging, sedimentation, and deposition can be treated as size-dependent. The number of bins may vary from 2 (fine and coarse particles; e.g., LOTOS-EUROS, Schaap et al., 2008) up to 24 or more. The model accuracy increases with size resolution (i.e., the number of sections), but satisfactory results are typically obtained already with 12 sections over the range 1 nm to 10 µm (Zhang et al., 1999; Devilliers et al., 2013). Both sectional and modal approaches have pros and cons that were evaluated in detail by Zhang et al. (1999).

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4.3.4 Internal mixture

It is impossible to describe the full complexity of aerosol chemical composition in 3-D numerical models. A common approximation is to assume that the percentage contribution of the individual compounds is the same for all particles within one mode or one section. This mixing state is then called the internal mixture. It is used in most aerosol modules.

4.3.5 External mixture

The opposite approximation would be that each mode or section consists of particles that may have different chemical composition. This state is then called the external mixture. The simulation of an aerosol population that is distributed both in size and chemical composition is challenging and, to date, 3-D simulations have typically included various approximations to simulate external mixtures (Jacobson et al., 1994; Kleeman and Cass, 2001; Oshima et al., 2009; Lu and Bowman, 2010; Riemer et al., 2009). A full discretization of both size and chemical composition is feasible (Dergaoui et al., 2013), but it has not yet been incorporated into 3-D models.

Models often include a combination of internal and external mixtures by simulating several overlapping modes (representing the external mixture) each having a different (internally mixed) chemical composition.

4.3.6 Treatment of secondary organic and inorganic aerosol

The chemical compounds found in aerosols can be differentiated between inorganic and organic species. The main inorganic compounds are nitrate, sulphate, ammonium, chloride and sodium. Calcium, magnesium, potassium and carbonate can be considered to represent alkaline dust. The partitioning of these species between gas and particle phase and their thermodynamic state in the particle phase (solid or liquid) can be calculated by solving the equations of thermodynamic equilibrium and minimizing

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the Gibbs energy of the system. The exact solution is computationally expensive (e.g., AIM2 – Wexler and Seinfeld, 1991) and has been seldom used in 3-D models. Solving the system via a set of selected equilibrium relationships that can be optimized according to the chemical regime of the system is the most widely used approach in 3-D modelling: such thermodynamic models of inorganic aerosols include for example EQUISOLV II (Jacobson, 1999), ISORROPIA, ISORROPIA II (Fountoukis and Nenes, 2007), MOSAIC (Zaveri et al., 2008), and PD-FiTE (Topping et al., 2009, 2012). The equilibrium between the gas phase and the particles is reached rapidly for fine particles but may take several minutes for coarse particles. Some air quality models account for this potential mass transfer limitation by implementing a dynamic approach, which can be limited to coarse particles for computational efficiency. Zhang et al. (2000) present a comparative evaluation of various inorganic thermodynamic aerosol models.

Carbonaceous components of aerosol particles include black carbon (BC, also referred to as elemental carbon or light-absorbing carbon) and organic compounds. BC is typically treated as entirely present in the particle phase and chemically inert. The number of organic species found on aerosol particles is huge and it is impossible to treat each single one in 3-D online coupled models. To solve this problem, similarly to gas-phase chemistry, surrogate species are introduced to represent organic aerosols (OA), which can be primary (POA) and secondary (SOA). Three main approaches are currently in use to model organic aerosols: the two-product Odum approach, the volatility basis set (VBS) and the molecule surrogate approaches. The two-product Odum approach is an empirical one that assumes the production of SOA from the oxidation of a VOC precursor by a given oxidant (i.e., OH, NO_3 or O_3) can be represented by two surrogate SOA semi-volatile species, which are represented by their mass stoichiometric coefficient and their gas/particle partitioning coefficient (which can be temperature-dependent) (Odum et al., 1996; Schell et al., 2001). No assumption is made about the molecular structure of those surrogate species, but it is implicitly assumed that they are hydrophobic and that their activity coefficients are constant. The VBS approach (Donahue et al., 2006) is also an empirical approach, but it differs from the previous

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one by its definition of surrogate SOA compounds, which also allows the treatment of additional processes such as chemical ageing and semi-volatile POA (Donahue et al., 2006). Surrogate SOA species are predefined according to a set of saturation vapour pressures (i.e., the VBS, which corresponds to a discretization of all the possible volatilities of SOA species). Then, chemical ageing within the particle may move a fraction of a compound to a less volatile compound. Also, semi-volatile POA can be treated with this formulation. The approach has been recently extended to account also for the degree of oxidation of SOA species (Donahue et al., 2011). A simplified version of such a scheme is described by Athanasopoulou et al. (2013).

The molecule surrogate approach is based on a mechanistic approach to organic aerosol formation by using surrogate SOA molecules that are determined from the experimental characterization of the chemical composition of organic aerosols (Pun et al., 2002, 2006). As for gas-phase mechanisms, uncertain parameters (stoichiometric coefficients and gas/particle partitioning constants) are specified following comparison with experimental data. This approach offers the advantage that SOA species can be either hydrophilic or hydrophobic (or both) and that the activity coefficients are not constant, but may evolve with the aerosol chemical composition. Semi-volatile POA can also be treated with this approach.

4.3.7 Treatment of chemical ageing of particles

Heterogeneous reactions and condensational growth during transport alter the mixing state of particles and contribute to aerosol ageing. The mixing state of soot particles is important for two reasons. Pure soot particles are hydrophobic. This means there is no hygroscopic growth when inhaled by humans and, as they are small, they can get deep into the lungs causing health problems. Ageing of pure soot particles occurs by condensation of gas phase compounds or by coagulation with particles of different chemical composition. If these compounds form a soluble shell around the soot core, those internally mixed particles become hygroscopic. Such particles, when getting inhaled, grow very rapidly and are deposited early in the respiratory tract. In addition,

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soluble shells alter the specific shortwave absorption of soot particles to higher values. The potential of soot particles to act as CCN or ice nuclei (IN) is also modified by the ageing process. The ageing of mineral dust particles changes their capability to act as IN or CCN. Therefore, the explicit treatment of the ageing process is an important process that should not be neglected. An example for models that treat soot ageing explicitly is MADEsoot (Riemer et al., 2003).

4.3.8 Aqueous-phase formation of aerosol species

The formation of inorganic and organic aerosols may also occur via aqueous chemical reactions and subsequent cloud droplet evaporation. Many 3-D models treat the oxidation of SO_2 to sulphate and NO_x to nitrate in clouds via homogenous and heterogeneous reactions, as those processes contribute significantly to sulphate and nitrate formation. Few 3-D models have included the formation of SOA in clouds so far, but simulations conducted to date suggest that this pathway could contribute significantly to SOA concentrations locally and on the order of 10 % on average (Chen et al., 2007; Carlton et al., 2008; Ervens et al., 2011; Couvidat et al., 2013).

4.3.9 Aerosol chemistry and thermodynamics

The chemical composition of aerosols is modified by exchange processes with the surrounding gaseous compounds. On the one hand, the chemical composition of particles determines their water content, while on the other hand, the water content determines the chemical composition of particles. In most 3-D models it is assumed that the particle phase is in a thermodynamic equilibrium with the gas phase. There are only a few models available to treat the thermodynamic equilibrium. Those are ISORROPIA II (Fountoukis and Nenes, 2007), MOSAIC (Zaveri et al., 2008), and PD-FiTE (Topping et al., 2009, 2012).

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4.3.10 Thermal diffusion

According to recent studies, a newly discovered aerosol phenomenon, called turbulent thermal diffusion (TTD), has a rather small but systematic contribution to the global distribution of coarse particles. TTD, first predicted theoretically by Elperin et al. (1996) and then found in laboratory experiments (Buchholz et al., 2004), entails the transport of particles against the temperature gradient and is more effective at low pressure. Sofiev et al. (2009b) showed that TTD is most likely responsible for the aerosol layer at tropopause height – a phenomenon not well explained so far. Their simulations with the SILAM model have shown that this regional effect to long-term average PM_{10} concentrations is of the order of 5–10 % in most areas, but in certain mountainous regions the concentrations are enhanced by 40 % (with respect to model runs without TTD) due to more efficient upward transport.

4.4 Model treatments of cloud properties

4.4.1 Bulk schemes

Regional models treat cloud properties to various degree. The most common approach is to use bulk schemes in which the moments of a given number of hydrometeors are predicted. With only one moment, the mass mixing ratio is predicted, then the number concentration has to be prescribed or parameterized and the size distribution is very much idealized. The simplest example of such a one-moment scheme is the Sundqvist (1978) scheme in which only the sum of cloud water and cloud ice is predicted and the distinction between cloud water and cloud ice is based only on temperature. In one standard NWP model used by several European weather services (Baldauf et al., 2011), clouds are represented by a bulk microphysics scheme, which describes different categories of cloud hydrometeors by size distribution functions, for which the mass mixing ratio is predicted and the number concentration is prescribed. Following Houze (1994), cloud droplets, raindrops, cloud ice, and snow are taken into account. Micro-

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physical processes are represented by transferring hydrometeors from one of those categories to another.

In order to consider the different freezing mechanisms and their dependence on the available ice nuclei, separate prognostic variables for cloud water and ice need to be solved. General circulation models (GCM) typically use one-moment schemes for predicting cloud water and ice separately (e.g. Lohmann and Roeckner, 1996). Regional models often solve additional prognostic equations for falling hydrometeors (rain, snow, graupel and sometimes hail) (e.g., Seifert and Beheng, 2006).

Two-moment schemes (e.g., Seifert and Beheng, 2006) predict the number concentrations of the hydrometeors in addition to their mass mixing ratios. Scientifically, they are superior to one-moment schemes because the nucleation of cloud droplets can be parameterized according to Koehler's theory (e.g., Abdul-Razzak et al., 1998) and can take the dependence of the aerosol number concentration into account. They can also account for the size-dependent sedimentation rate (Spichtinger and Gierens, 2009).

Bin schemes (see Khain et al., 2008 as an example) are best suited to resolve the cloud droplet size distribution, but they are computationally more expensive and are usually only used for research applications.

4.4.2 Processes taken into account

Condensation/deposition of water vapour

Condensation is treated with a saturation adjustment scheme, which means that all the water vapour above 100 % relative humidity is converted into cloud water based on the assumption that sufficient CCN are available to deplete the supersaturation. This assumption is justified for water clouds, where the supersaturation with respect to water is at most 1 %. However it is questionable for ice clouds, where supersaturation with respect to ice can reach 70 %. Therefore, GCMs and regional climate models (RCMs) have started to abandon the saturation adjustment scheme for cirrus clouds

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and allow supersaturation with respect to ice (Lohmann and Kärcher, 2002; Liu et al., 2007; Tompkins et al., 2007; Gettelman et al., 2010; Salzmann et al., 2010).

In two-moment schemes, the number of activated aerosol particles determines the number of nucleated cloud droplets. If parameterizations based on the Köhler theory (Abdul-Razzak and Ghan, 2002; Nenes and Seinfeld, 2003) are used in online coupled models, it makes sense to abandon the saturation adjustment scheme and instead solve the droplet growth equation. More details as well as the parameterization of ice nucleation are given in Sect. 4.5.

Formation of precipitation

The first microphysical scheme was developed by Kessler (1969). It distinguishes between cloud water with small drop sizes (5–30 µm) having negligible velocity and rain drops that reach the surface within one model time step. The conversion from cloud water to rain, the autoconversion rate, depends only on cloud water content and starts once the critical cloud water content is exceeded. A similar scheme for the ice phase (Lin et al., 1983) is widely used in RCM and NWP models.

If cloud water and cloud ice are predicted as separate prognostic variables, the Bergeron–Findeisen process can be parameterized to describe the growth of ice crystals at the expense of water droplets due to the lower vapour pressure over ice in mixed-phase clouds. It depends on the updraft velocity inside the cloud and the number concentration of ice crystals (e.g., Storelvmo et al., 2008).

4.5 Aerosol-cloud interactions and processes

4.5.1 Aerosol-cloud interactions in online models

Aerosols are a necessary condition for cloud formation and influence cloud microphysical and physical properties as well as precipitation release. Hence, all online models have cloud schemes that to some extent represent the effect of aerosols on clouds

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(e.g. RegCM, Giorgi et al., 1993; some versions of MetUM, Birch et al., 2012). If no prognostic aerosol number and mass concentrations are coupled to the cloud scheme the aerosols are implicitly assumed in the parameterizations of the cloud scheme. One example is the widely used diagnostic calculation of CCN number concentration (often assumed to be the cloud droplet number concentration) in the warm phase: $n_d = cs^k$, where s is the supersaturation and c and k are empirically derived coefficients that differ for different aerosol loadings. The cloud droplet number is often used in parameterizations of the cloud droplet effective radius, which is a basic parameter for parameterizing cloud radiation interactions. The values of c correspond to the CCN concentration at 1 % supersaturation, while k is a tuning coefficient. Information on aerosol number, size and composition is contained within c and k. Accordingly, c retains large values (e.g., 3500 cm^{-3}) in continental and polluted locations, but small values (e.g., 100 cm^{-3}) at remote marine locations (Hegg and Hobbs, 1992). The supersaturation field is, however, also strongly influenced by the aerosol concentration, and the parameterization of s likewise implicitly contains information on aerosol number, size and composition.

The CCN concentration in the above example will likely not reflect the strong temporal and spatial variations of the real atmospheric aerosol concentrations. In addition, the implicitly assumed aerosol properties may be inconsistent and give rise to biases in the cloud radiation interactions as well as in cloud water and cloud ice content.

4.5.2 Aerosol-cloud interactions in online models with prognostic aerosols

Several European online models take into account the influence of aerosols on cloud properties via an explicit treatment of aerosol concentrations. In Enviro-HIRLAM (Korsholm, 2009; Baklanov, 2008) the aerosol indirect effects have been implemented in a one-moment cloud scheme. In WRF-Chem (Gustafson et al., 2007; Chapman et al., 2009; Yang et al., 2012) several double moment microphysics modules are coupled to prognostic aerosol properties. In RAMS/ICLAMS (Solomos et al., 2011) as well as in COSMO-ART (Bangert et al., 2011) and MetUM (Collins et al., 2011) a detailed two-

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moment microphysical scheme is also directly coupled with prognostic aerosol properties.

In all models, cloud–radiation interactions are parameterized via the effective cloud droplet radius, employed in the respective radiation schemes. The effective cloud droplet radius is dependent on cloud droplet number concentration as well as cloud mass in the warm phase of the clouds. As the cloud droplet number increases for a constant liquid water content, the effective radius decreases and clouds become more reflective to incoming shortwave radiation.

In Enviro-HIRLAM, activation of anthropogenic aerosol into cloud droplets is treated using the empirical relation of Boucher and Lohmann (1995) relating cloud droplet number to sulphate mass concentration. A constant natural (background) cloud droplet number contribution is assumed. In WRF-Chem the cloud droplet number concentration is treated prognostically in several two-moment cloud schemes (Lin et al., 1983; Morrison et al., 2005; Morrison and Gettleman, 2008; Ghan et al., 1997). The modelled aerosol size distribution and composition affects the activation process, which is calculated for both number and mass fractions following Abdul-Razzak and Ghan (2002). Evaporation of cloud droplets and resuspension of aerosols to an interstitial mode is also accounted for (see Gustafson et al., 2007; Chapman et al., 2009; Yang et al., 2009 for an overview). In RAMS/ICLAMS the cloud droplet number concentration is calculated based on a prognostic representation of aerosol size and chemical composition within the framework of an ascending adiabatic cloud parcel (Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005). This scheme can be extended to include adsorption activation from insoluble CCN following Kumar et al. (2009). or by the consideration of effect of entrainment on activation (see Barahona and Nenes, 2007).

The supersaturation needed for activating a CCN is determined by the modified Köhler theory that takes the effects of surfactant and slightly soluble species into account. The effects of CCN on cloud droplet activation can also be considered following Barahona et al. (2010). In MetUM, activation follows an empirical approach to couple aerosol mass and activated number concentration, while COSMO-ART employs an

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aerosol composition dependent calculation of supersaturation and Köhler theory (Ghan et al., 1997) to calculate aerosol activation rate.

In the STRACO cloud scheme (Soft TRAnsition and Condensation: Sass, 2002) used in Enviro-HIRLAM, the default empirically-based Sundqvist-type autoconversion (Sundqvist, 1988) has been replaced by a relationship from Rasch and Kristjansson (1998). Hereby, the autoconversion process and hence the onset of precipitation becomes dependent on cloud droplet number. In WRF-Chem a physically based autoconversion threshold function (Liu et al., 2005) has been implemented in the cloud scheme to represent the effect of cloud droplet number on the conversion of cloud water to rain. In RAMS/ICLAMS the two-moment scheme of Meyers et al. (1997) is used for predicting the mixing ratio and number concentration of seven hydrometeors (cloud, rain, pristine ice, snow, aggregates, graupel and hail). The scheme considers autoconversion of cloud droplets to rain drops due to collision and coalescence, breakup of rain droplets, diagnosis of ice crystal habit, evaporation and melting of each species and shedding. The autoconversion scheme used in MetUM relies on the scheme by Khairoutdinov and Kogan (2000) in which the precipitation development depends on cloud droplet mean volume radius, droplet number and liquid water content. In COSMO-ART the conversion of cloud water to precipitation follows Seifert and Beheng (2001) who assumes the shape of the droplet spectrum to be a generalized gamma distribution.

The one-moment cloud schemes used in Enviro-HIRLAM are typical of operational models with strict execution time requirements. As the cloud droplet number increases and the effective radius decreases with an increased aerosol loading, the microphysical processes are also affected. For instance, the changed surface area of droplets yields to alteration of evaporation and condensation. However, when diagnosing cloud droplet number concentration these effects are not accounted for. The importance of such effects and the associated changes in cloud dynamics has been discussed by several authors (see e.g., Xue and Feingold, 2006; Jiang et al., 2006). Recently, it has been suggested that for clouds with liquid water path less than about $50\,g\,m^{-2}$ the evaporation/condensation effect is of greater importance in controlling the liquid water

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path than the precipitation suppression effect (Lee and Penner, 2011). Therefore, it might be misleading to represent aerosol–cloud interactions in terms of the effect on autoconversion only.

Full microphysical bin-resolved descriptions of cloud and aerosol microphysics are, however, still computationally expensive and generally not feasible for operational forecasting. The influences of aerosols on clouds must be thence parameterized. A problem arising in this context is that most online models rely on empirical relationships for representing the processes where aerosols and clouds interact. Examples of such empirical parameterizations include the relation between sulphate mass and cloud droplet number concentration (Boucher and Lohmann, 1995; Jones et al., 1994) or the relation between aerosol number concentration near cloud base and cloud droplet number concentration by Martin et al. (1994). In order to simulate the interaction processes in short-range models, it is important that the parameterizations are based on the relevant coupling processes and that tuning affects only parameters that are not influenced by the coupling. Physically-based schemes that explicitly resolve the activation of CCN into cloud droplets (e.g. Abdul Razak and Ghan, 2002; Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005; Barahona et al., 2010) are expected to improve the representation of these processes in regional models.

4.5.3 Parameterization of ice nucleation

Aerosol effects on ice clouds are even more uncertain than aerosol effects on water clouds. A small subset of aerosols, such as mineral dust, acts as ice nuclei (IN) and determine the formation of the ice phase in clouds. The importance of other aerosols (biological particles, black carbon, organic carbon or crystalline ammonium sulphate) acting as IN is still a matter of debate. While biological particles have been found to nucleate ice at the warmest temperatures, their concentrations in the atmosphere seem to be too low to have a global impact (Hoose et al., 2010a,b; Sesartic et al., 2012). The opposite is true for black and organic carbon as they may nucleate ice at only slightly warmer temperatures than needed for homogeneous freezing.

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Cirrus clouds form at temperatures below −35 °C. Here homogeneous freezing of solution droplets prevails. Heterogeneous freezing on ice nuclei seems to be of minor importance but can be important in determining the maximum supersaturation. Parameterizations of cirrus schemes that consider the competition between homogeneous and heterogeneous nucleation have been developed by Kärcher et al. (2006), Barahona and Nenes (2009), Gettelman et al. (2010), Salzmann et al. (2010), and Wang and Penner (2010).

At temperatures above −35 °C, ice forms heterogeneously in the mixed-phase cloud regime. Most models describe ice formation in mixed-phase clouds with empirical schemes (e.g., Lohmann and Diehl, 2006; Phillipps et al., 2008; DeMott et al., 2010). In order to consider which aerosols act as IN at a given temperature, laboratory data are used. As the ice nucleating properties of BC are still very uncertain, the potential anthropogenic effect of BC on ice clouds is also questionable. There are two possibilities. On the one hand, more BC aerosols cause a faster glaciation of supercooled liquid clouds inducing faster precipitation and shorter cloud lifetime. This counteracts the warm indirect aerosol effects and will reduce the total anthropogenic aerosol effect (Lohmann, 2002). On the other hand, if anthropogenic BC is predominantly coated with soluble species, this may reduce its ability to act as an IN and works in the opposite way (Storelvmo et al., 2008; Hoose et al., 2008). Which of these effects dominates remains an open question. The newest and most physical approach is to parameterize heterogeneous freezing in mixed-phase clouds based on classical nucleation theory (Hoose et al., 2010b).

4.6 Radiation schemes in coupled models

Online-coupling imposes additional requirements on the setup and implementation of radiation modelling schemes, particularly when gas and aerosol feedbacks are explicitly considered. Most of these requirements reflect the need to maintain physical and numerical consistency between the various modules and computational schemes of

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the model, against the increased frequency of interaction (typically of the order of a few dynamical time steps) and the multitude of simulated effects.

4.6.1 Radiative effects of gases and aerosols

Trace gases such as ozone, nitrogen oxides and methane absorb incoming shortwave radiation and thereby modify the radiation balance at the ground and photolysis rates. The main key parameter determining the absorption of radiation by a particular gas is its concentration profile. Aerosol particles absorb, scatter and re-emit both short and long-wave radiation thus directly affecting the surface radiation balance, heating rates in the atmosphere (direct aerosol radiative effect) and, in the case of shortwave radiation, photolysis frequencies and also visibility. Key species to be considered are water attached to aerosol particles, sulphate, nitrate and most organic compounds, which mostly result in a cooling of the atmosphere, and BC, iron, aluminium, and polycyclic/nitrated aromatic compounds, which warm the air by absorbing solar and thermal-infrared radiation.

The role of cloud droplets is also important as optical properties of the droplet ensemble are influenced by the size distribution and composition of the aerosol particles acting as CCN. In the case of particles, not only the mass concentration but also their composition, the size distribution of both aerosol particles and cloud droplets and the mixing state (internal or external mixture) have a strong effect on the interaction with solar radiation.

All direct radiative effects will result in the development of semi-direct effects like changes in thermal stability, cloudiness, etc. Although the inclusion of semi-direct effects does not generally require the explicit incorporation of extra processes in the models, the radiation modules need to be able to adequately resolve the atmospheric radiation fluxes associated with each process.

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4.6.2 Implementation considerations

Although online coupled models mostly consider, at least, the aerosol direct effect, the level of details for considering this effect differs strongly between models. With respect to the inclusion of the radiative effect of trace gases and cloud droplets there are also major differences between the models.

Some of the online coupled models follow the approach of simulating aerosol radiative properties by calculating complex refractive indices and extinction coefficients of particulate matter (PM) and cloud components as a function of size distributions and chemical composition for a specific mixing state by Mie calculations during runtime. In order to speed up costly calculations of radiative feedbacks, some other radiation modules use externally stored data in the form of a pre-computed parameter cache (e.g., tabulated results of a priori Mie calculations or from the OPAC – Optical Properties of Aerosols and Clouds – software library module).

Even in the case when optical properties of aerosol particles and droplets are directly calculated, the result may still depend on the representation of size distributions (modal; number of moments/sectional; number of bins), the considered compounds, and on assumptions on the mixing state of the particles. Besides these differences in the description of the optical properties of trace gases, aerosol particles, and cloud droplets, which are input to the radiation scheme, the degree of detail of the radiation scheme may also depend on the level of accuracy of the radiation scheme itself (2-stream or higher order, spectral resolution).

The introduction of such advanced radiation schemes in online coupled models can often require significant development effort, due to both the inherent limitations of traditional radiation modules as well as incompatible representations of chemical speciation and thermodynamic properties between CTMs and radiation modules. It also imposes a significant computational burden, by requiring that radiation modules are invoked typically at every (or every few) dynamical step(s) of the model. Inevitably, modellers have to resort to simplifications in both the selection of parameterisations and simu-

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lated effects, as well as the number of particle size bins and mixing states. Even more stringent limits have to be imposed on the number of simulated spectral bands which are required for an accurate account of the full range of radiative effects of complex PM mixtures. Nielsen et al. (2011) have investigated ways to reduce the computational footprint of shortwave spectral calculations by substituting 2-stream calculations for averages of aerosol optical properties weighted over the entire solar spectrum. In any case, the effect of any simplification or speed-up of the radiation modules should be thoroughly evaluated through extensive validation studies.

4.6.3 Review of radiation schemes in online-coupled models

Online coupled models can, in theory, enable the full range of radiative feedbacks but in practice only some include any. Interactions between gas-phase chemistry and radiation are frequently introduced via an online coupling of the dynamical core and radiation modules with the photolysis module.

Only a few models account for the effect of variable trace gas concentrations on solar and longwave radiation. Absorption by variable tropospheric ozone and CO_2 is included in COSMO-ART, Enviro-HIRLAM, and in the RRTMG and CAM radiation parameterization of WRF-Chem, while RAMS/ICLAMS includes the option of using simulated ozone in the radiation schemes (see Table 8).

The complexity of the treatment of the effect of simulated aerosol concentrations on shortwave and longwave radiation fluxes differs strongly between the models. For example, MCCM considers only the simulated total mass of dry aerosol in each layer in combination with a "typical" mass extinction coefficient and the water attached to the aerosol in order to calculate the extinction of shortwave radiation. Different chemical species, which include BC, organic mass (OM), water, and various ionic species, such as sulphate and nitrate can be considered in COSMO-ART, WRF-Chem, and Enviro-HIRLAM. Within WRF-Chem an aerosol optical property module (Barnard et al., 2010) treats bulk, modal, and sectional aerosol size distributions using a similar methodology for refractive indices and multiple mixing rules to prepare 3-D distributions of aerosol

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optical thickness, single scattering albedo, and asymmetry parameters that are passed into some of the shortwave and longwave radiation schemes available. While soot is considered as externally mixed in COSMO-ART, all compounds are assumed to be internally mixed in WRF-Chem. WRF-Chem offers the option of runtime Mie calculations for the optical parameters (using bulk, modal, or sectional aerosol modules) as well as BOLCHEM (Russo et al., 2010) and COSMO-MUSCAT (Heinold et al., 2008) for dust aerosols. In COSMO-ART actual optical parameters are calculated based on these tabulated values derived from aerosol distributions of a previous COSMO-ART run and the actually simulated aerosol masses of each mode (Vogel et al., 2009). A slightly different approach is chosen in MEMO/MARS-aero, where radiative effects of aerosol particles are introduced using the OPAC software library (d'Almeida et al., 1991). OPAC defines a dataset of typical clouds and internally mixed aerosol components, which are externally mixed to calculate the optical properties from the concentration fields of simulated PM compositions. Within the cloud droplets the effects of dissolved sulphate, speciated PM and trace gases are taken into account by Enviro-HIRLAM.

4.7 Emissions and deposition and their dependence on meteorology

Emissions are essential inputs for CTMs. Uncertainties in emissions and emission parameterizations rank among the largest uncertainties in air quality simulations. In the context of online coupled modelling, the most interesting emissions are those which depend on meteorology as these could potentially be treated more accurately and consistently than in offline models. This section therefore focuses on this type of emissions. Chemical species are ultimately removed from the atmosphere by dry and wet deposition which are strongly driven by meteorology and therefore almost always calculated online. However, similarly to emissions, meteorological dependencies are sometimes neglected or simplified, for example when constant dry deposition rates are a priori prescribed.

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4.7.1 Emissions

Inventories of anthropogenic emissions typically contain annual total mass emissions of the most important species and compound families such as NO_x, SO_x, methane and other VOCs, ammonia, and some PM species (e.g. organic matter, elemental carbon, trace metals). The most commonly used emission inventories in Europe are those of the European Monitoring and Evaluation Programme (EMEP – http://www.ceip.at/) and the inventory developed within the EU FP7 MACC project (Monitoring Atmospheric Composition and Climate; Kuenen et al., 2011). Annual emissions are translated into emissions for a given month of the year on a given day of the week and a given hour of the day by using category-specific time factors based on activity data. Recently, meteorological dependencies of anthropogenic emissions have started to be taken into account in some models, like the increased energy demand under cold periods (residential heating). Agricultural emissions of ammonia and dust are typically guided by meteorology, which is at present not taken into account other than via calendars. A more interactive treatment of these types of meteorology-depending emissions would be desirable.

Natural emissions are closely related to meteorology, and are in general already calculated online even in offline models using the meteorological input driving the CTM. Sea spray is the dominant aerosol source over the oceans and therefore, its proper quantification is highly relevant for a coupled model. Sea salt emissions depend on (at least) wind speed and sea water temperature, but there is also evidence of a dependency on wave state and organic matter concentrations (see De Leeuw et al., 2011, for a recent overview). The fluxes and composition of the smallest particles are especially uncertain. In addition to direct radiative effects (Lundgren et al., 2012), sea-salt aerosols may feed back on meteorology by acting as CCNs, their activity being dependent on the organic fraction contributed by phytoplankton (Ovadnevaite et al., 2011). In addition, phytoplankton is a source of dimethyl sulphide (DMS) and a minor source of

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isoprene (Guenther et al., 1995). All models include sea-salt, but DMS and the organic fraction are not always taken into account, as they are more uncertain.

Windblown dust refers to particles from a broad range of sources. Due to their direct relationship with meteorology, windblown dust emissions must be calculated online. Natural emissions of dust, for example, from deserts, depend on wind speed and soil characteristics (type, vegetation, texture, wetness). If such relations are nonlinear, online models have a clear advantage. Some parameterizations account for the dependence of the size distribution of the vertically emitted particles on the size of the saltation particles (Alfaro and Gomez, 2001; Shao, 2001), while others do not consider this dependency (Marticorena and Bergametti, 1995). Road resuspension may be an important source in some areas depending on traffic intensities, use of studded tires, the amount of dust on a road, sanding activities, soil moisture and rain and snow. Also agricultural activities contribute to windblown dust, depending on arable soil land fractions, timing of activities and their intensity, translations of activity to dust release, as well as rain, snow, and temperature. Agricultural emissions are taken into account in only a few models and even in those models, they are crudely calculated and poorly validated. For a more detailed description of windblown dust emissions see the report of Schaap et al. (2009) and references therein.

Emissions of biogenic volatile organic compounds (BVOC) like isoprene and terpenes are strong functions of meteorological conditions. Measurements on individual plant species demonstrated that temperature and photosynthetically active radiation (PAR) are the key driving variables for these emissions (e.g., Tingey et al., 1980; Guenther et al., 1991; Staudt et al., 1997). Most models therefore calculate these emissions online based usually on the MEGAN scheme (Model of Emissions of Gases and Aerosols from Nature; Guenther et al., 1993, 1995, 2006, 2012) or variations thereof (Vogel et al., 1995), but effects of stress like high ozone concentrations or drought are usually not taken into account. BVOC emissions contribute to ozone formation and can also give a significant contribution to the formation of SOA, which in turn may affect radiation and hence feedback on BVOC emissions in an online coupled model.

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Other biogenic emissions of potential relevance for air quality simulations and chemistry–meteorology interactions are NO emissions from soils (Yienger and Levy II, 1995) and emissions of bacteria, fungal spores and pollen which have been reported to act as efficient ice nuclei (e.g., Hoose et al., 2010a,b; Pummer et al., 2012). Lightning is an important natural source of NO_x in the free troposphere, but it is still often neglected in mesoscale models. Online coupled models with access to convective mass fluxes or cloud fields calculated by the core meteorological model, however, would be perfectly suited to simulate this source (Tost et al., 2007). In contrast to most gases that are consistently deposited, NH_3 fluxes over fertilized agricultural lands and grazed grasslands are bi-directional, with both deposition and emission occurring in parallel (Sutton et al., 1998; Zhang et al., 2008). Advanced resistance models accounting for capacitance of leaf surfaces have been developed to simulate the bi-directional NH_3 exchange (e.g., Nemitz et al., 2001; Wu et al., 2009) and incorporated into WRF-CMAQ. Wichink Kruit et al. (2012) showed that by including bi-directional exchange in the LOTOS-EUROS model, ammonia concentrations were increased almost everywhere in Europe and nitrogen deposition was shifted away from agricultural areas towards large natural areas and remote regions. Pollen emissions are dependent on meteorology and season (e.g., Sofiev et al., 2009a) and have an impact on visibility. Pollen emissions are included in Enviro-HIRLAM (birch) and COSMO-ART (birch and grass).

The occurrence of forest fires is dependent on drought and their spread is partly determined by wind conditions. However, the start of a forest fire is a random process, which can be initiated by lightning but also by human beings. Therefore, fire emissions cannot be calculated as being purely dependent on meteorological conditions but, for simulations of the past, are based on satellite observations of wildfires. These wildfires emit large amounts of CO_2, CO, and VOCs as well as primary PM (EC, OC) and have an impact on ozone and PM concentrations, as well as on visibility.

Most of the problems and uncertainties associated with emissions of gaseous and particulate pollutants of anthropogenic and natural origin are not specific to online coupled models. However, due to the important role of aerosols in chemistry–meteorology

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feedback processes, the main efforts of the online modelling community should be devoted to improved descriptions of primary aerosol emissions and of emissions of the most relevant gaseous aerosol precursors including ammonia, NO_x and biogenic VOCs. For any type of emitted primary aerosol, a better physico-chemical characterization in terms of number and mass size distributions, hygroscopicity, internal versus external mixture, would be desirable as number concentrations and size distribution have a major impact on their cloud forming properties and radiation effects, rather than their mass concentrations. Such inventories are currently being developed (Visschedijk and Denier van der Gon, 2011). Wind-blown dust emissions from dry soils and resuspension of particles are still poorly understood and described in models. While not being a major health concern, mineralogical particles play an important role in cloud formation notably by acting as ice condensation nuclei (Hoose et al., 2008, 2010b; Gettelmann et al., 2010). A better meteorology-dependent parameterization of agricultural particle emissions would be also advisable as they contribute significantly in some areas. Regarding emissions of gaseous aerosol precursors, model improvements should particularly focus on the following issues:

i Ammonia emissions should be described in a more interactive way taking meteorology and agricultural practices into account and considering bi-directional exchange.

ii A better quantification of BVOC emission factors for the major tree species in Europe and better geographical maps of their spatial distribution are needed to reduce the large spread in BVOC emissions between models.

iii The contribution of anthropogenic VOC emissions to SOA formation appears to be rather minor on both the global (Henze et al., 2008) and European (Simpson et al., 2007; Szidat et al., 2006) scales, but primary emissions of organic particles from fossil and wood burning may make a significant contribution to total carbon within Europe (Simpson et al., 2007) and thus, should receive more attention.

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4.7.2 Wet and dry deposition and sedimentation

Wet and dry deposition are the only possible pathways to remove material previously released into, and processed in, the atmosphere. Both processes are directly driven by meteorology and thus, online coupled models have a high potential to describe these processes more accurately, e.g. because all meteorological fields are directly accessible at high temporal resolution.

Dry deposition has mainly an effect on surface concentrations, except for heavy particles (diameters larger than about 10 µm) which fall out relatively quickly. Dry deposition is directly driven by meteorology (temperature, humidity, snow/wet surface, wind speed; e.g., Wesely, 1989; Zhang et al., 2001, 2003; Seinfeld and Pandis, 2003) and indirectly through soil moisture.

Wet deposition is an efficient removal mechanism for many gases and aerosols depending on the scavenging ratio and collection efficiency for the species. In contrast to dry deposition at the surface, it impacts gases and aerosols over a larger vertical extent from the surface to cloud top. It is often simulated as two separate processes, in-cloud and below-cloud scavenging, and depends on the presence of clouds, cloud and droplet properties and the precipitation rate. Treatments of wet scavenging and cloud processing are often highly simplified and vary strongly between models (Gong et al., 2011). A detailed scheme was recently developed for COSMO-ART taking full advantage of the access to microphysical tendencies (condensation, evaporation, autoconversion, etc.) simulated by the meteorological core model which would not easily be possible in an offline model (Knote and Brunner, 2013).

4.8 Chains/loops of interactions and other feedback mechanisms

The above described interaction mechanisms between aerosols, chemical and meteorological processes depend on each other and can as well interact with each other. The range of interactions can be much broader and hence cannot be fully covered by the simplified classification of aerosol feedbacks of the direct, semi-direct, first and

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second indirect effects. For example, the following effects of aerosol particles on meteorology and climate can be distinguished (Jacobson, 2002): (i) self-feedback effect, (ii) photochemistry effect, (iii) smudge-pot effect, (iv) daytime stability effect, (v) particle effect through surface albedo, (vi) particle effect through large-scale meteorology, (vii) indirect effect, (viii) semi-direct effect, (ix) BC-low-cloud-positive feedback loop. It is important to stress that many of the above-mentioned mechanisms are still uncertain, but their accurate description would require an online chemistry–meteorology integration together with the adequate resolution of meso-meteorological phenomena and a detailed ABL structure.

Besides, on a more general level as mentioned in Sect. 2, different chains/loops of interactions take place and should be properly simulated in OCMC models. Some examples of these chains of interactions are listed below:

- Aerosol → radiation → photolysis → chemistry;
- Temperature gradients → turbulence → surface concentrations, boundary layer outflow/inflow;
- Aerosol → cloud optical depth through influence of droplet number on mean droplet size → initiation of precipitation;
- Aerosol absorption of sunlight → cloud liquid water → cloud optical depth.

One example of such interactions chain of impacts from temperature on concentrations and vice versa was presented schematically in Fig. 2.

Zhang et al. (2010a) analysed with the online WRF-Chem model such a "chain effect" over the continental US and observed enhanced stability as a result of the warming caused by BC in the ABL and the cooling at the surface resulting from reduced solar radiation. Reduced ABL height indicates a more stable ABL and can thus further exacerbate air pollution over areas where air pollution is already severe. Similar chain effects were found in applications of WRF-Chem over East Asia, Europe, and globally (Zhang et al., 2012c, 2013).

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Due to strong non-linearity, space and time inhomogeneity of different interacting mechanisms, such chains and loops of interactions can be reproduced only using fully online integration of aerosol dynamics, chemistry and meteorological processes resolving/describing them together at the same timestep. The online access approach for coupling chemistry and meteorology models is very limited in its ability to produce such chain effects.

Besides, several additional feedback mechanisms of chemistry–meteorology interactions were not considered in the previous sections. The number of interactions between aerosols, gases and components of the Earth system is large (Fig. 3) and some of these interactions have been described in previous sections. The paragraphs below deal with the remaining mechanisms.

Light absorbing particles such as BC and dust affect climate not only by absorbing solar radiation but also changing snow albedo (Warren and Wiscombe, 1980, 1985; Painter et al., 2007). Dirty snow absorbs more radiation, thus heating the surface and warming the air even more. This warming initiates a positive feedback further reducing snow depth and surface albedo (Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2009). Through enhancing downwelling thermal infrared radiation, airborne BC influences the ground snow cover by increasing melting and sublimation of snow at night.

Absorption of solar radiation by BC and dust also perturbs the atmospheric temperature structure and thus affects clouds as well. By increasing atmospheric stability and decreasing relative humidity, BC is responsible for reducing the low-cloud cover, and by absorbing the enhanced sunlight, it warms the atmosphere further reducing cloud cover (Jacobson, 2002). While absorbing aerosols through the semi-direct effect contribute to cloud evaporation, their presence below the cloud level can enhance vertical motions by their heating effect and increased liquid water path and cloud cover. Koch and Del Genio (2010) found that the effects of absorbing aerosols could be a cooling caused by their effect on cloud cover and the cooling due to BC could be as strong as its warming direct effects.

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Intensification of air stability due to aerosol particles reduces surface winds and emission of particles and gas precursors that are wind dependent. The reduction of emissions due to thermal IR absorption is called the "smudge pot effect"; when the effect is due to both scattering and absorption of the solar radiation it is coined the "daytime stability effect" (Jacobson, 2002). Furthermore the local effect of aerosol particles on temperature and therefore on local air pressure, relative humidity, clouds and winds can influence the large-scale temperatures altering the thermal pressure systems and jet streams (Jacobson, 2002).

Moreover, aerosols can influence gas concentrations in the atmosphere through their direct interactions or by changing the actinic fluxes of ultraviolet radiation, and thus, accelerating or inhibiting photochemical reactions (Dickerson et al., 1997). Gases also interact with solar and IR-radiation to influence heating rates (photochemistry effect) (Jacobson, 2002). Among gaseous pollutants, tropospheric ozone is known to contribute to both AQ degradation and atmospheric warming. At high concentrations it can damage plant tissues resulting in a reduction of agricultural crop yields and forest tree growth (Turner et al., 1974; Krupa et al., 2000). Besides its direct effect as a warming GHG, ozone also generates an "indirect effect" onto the climate system through feedbacks with the carbon cycle. High ozone or carbon dioxide concentrations can cause stomatal closure, thus, limiting the uptake of the other gases. If ozone damages can be limited by increasing CO_2 concentrations, higher ozone concentrations can act on the photosynthesis, reducing CO_2 uptake and plant productivity, hence suppressing an important carbon sink. As a result carbon dioxide accumulates in the atmosphere and at global scale this effect can contribute more than the direct forcing due to ozone increase (Sitch et al., 2007).

The inclusion of the interactions depicted in Fig. 3 would allow considering the effects of the most important feedback mechanisms. Most of the regional models of Table 1 contain the aerosol direct effect on radiation, but only some of them the aerosol indirect effects (blue lines in Fig. 3). The chain of interactions from emissions (including wind-driven natural emissions calculated online), gas phase chemistry, formation of

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new particles, aerosol–cloud coupling and precipitation, and interactions with radiation (blue and green lines in the figure) is treated in the regional COSMO-ART model (Bangert et al., 2011; Knote et al., 2011). A similar degree of complexity in the realisation of feedback mechanism chain is achieved also in WRF-Chem (Gustafson et al., 2007; Chapman et al., 2009; Grell et al., 2011; Forkel et al., 2012), ICLAMS (Solomos et al., 2011) and Enviro-HIRLAM (Korsholm et al., 2008). In the last years, regional models have undergone significant developments in the direction of online coupling and although including a reduced number of feedback mechanisms, interesting studies were also conducted using COSMO-CLM (Zubler et al., 2011a,b), MESO-NH (Chaboureau et al., 2011), COSMO-ART (Stanelle et al., 2010), COSMO-LM-MUSCAT (Heinold et al., 2007), and NMMB/BSC-Dust (Perez et al., 2011).

An important feedback mechanism (red lines in Fig. 3) is linked to the light absorbing aerosol – albedo effect. It has been evaluated at regional scale using the WRF-Chem and WRF-RCM models (Qian et al., 2009) applied to the western United States. The study showed that changes in snow albedo due to BC deposition could significantly change regional climate and the hydrological cycle. Not only BC but also mineral dust deposition can reduce snow albedo and shorten the snow cover duration with feedbacks on climate at regional scale (Krinner et al., 2006). Thus, inclusion of BC and dust effects on snow albedo in regional models is desirable. Vegetation dynamics and ecosystem biogeochemistry (black lines in Fig. 3) can have positive feedbacks on temperature also at regional scale as evaluated using the RCA-GUESS model over Europe (Smith et al., 2011), as they can influence surface albedo and also emissions. Aerosols can influence the ocean biogeochemistry, the biosphere and the carbon cycle through other feedbacks. However, the level of understanding of many of these feedbacks is still low and their inclusion in models is still in an early phase (Carslaw et al., 2010).

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5 Numerical and computational aspects

With the increase of computational resources, more complex numerical models are becoming feasible, and an increase of the spatial resolution is affordable. Consequently, integrated meteorology-air quality models are experiencing closer attention in Europe. Key points in such models are the numerical schemes with especially those implemented for the transport of chemical species, the treatment of the coupling or integration between meteorology and chemistry, the role of initial and boundary conditions and the efficient performance of the system in a specific High Performance Computing (HPC) environment. This chapter addresses these issues with a final section discussing the state of development of data assimilation in chemical models and more specifically in online European models.

5.1 Numerical methods: advection schemes, mass consistency issues and other specifics

A number of different numerical techniques have been used and proposed for the transport of chemical tracers in online models in Europe. Some of them are able to maintain consistency of the numerical methods applied for both meteorological and chemical tracers (e.g., BOLCHEM, COSMO-ART, M-SYS, NMMB/BSC-CTM, REMOTE, WRF-Chem, some configurations of Enviro-HIRLAM, ICLAMS, AQUM), while others apply different transport schemes for meteorology and chemistry species, partly because the transport requirements for chemical species are stronger than those for hydrometeors in NWP (e.g., WRF-CMAQ, RACMO2/LOTOS-EUROS, some configurations of Enviro-HIRLAM). This may be a relevant deficiency when explicitly treating aqueous phase chemistry.

Rasch and Williamson (1990) listed the following desirable properties for transport schemes: accuracy, stability, computational efficiency, transportability, locality, conservation, and shape-preservation. The last two are of particular interest in chemistry modelling. Since Rasch and Williamson (1990), a number of additional desired proper-

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ties have emerged in the literature, with one of particular importance here: the so-called wind-mass consistency property (e.g., Jöckel et al., 2001). There is, however, one additional property not much discussed until recently, which is arguably the single most important desired numerical property for both off- and online transport of chemical tracers: preventing numerical mixing/unmixing (Lauritzen and Thuburn, 2011).

We briefly discuss below some of the above listed properties having particular relevance for chemical online modelling.

5.1.1 Conservation issues

Formal – or inherent – mass conservation of a numerical scheme can only be obtained if one solves – in one way or the other – the volume density equation for each chemical compound, i:

$$\frac{\partial \rho_i}{\partial t} = -\nabla \cdot \rho_i \boldsymbol{V} + S_i + D_i \qquad \text{or} \qquad \frac{\mathrm{d}\rho_i}{\mathrm{d}t} = \rho_i \nabla \cdot \boldsymbol{V} + S_i + D_i \tag{1}$$

where ρ_i is the volume density, $\boldsymbol{V}$ the velocity, while S_i represents sources/sinks (i.e. chemical reactions/emissions/deposition etc) and D_i turbulent diffusion/mixing. Just solving the corresponding equation for the mixing ratio, $q_i = \rho_i/\rho_\mathrm{d}$ (with ρ_d the actual dry density of air)

$$\frac{\partial q_i}{\partial t} = -q_i \nabla \boldsymbol{V} + S_i + D_i \qquad \text{or} \qquad \frac{\mathrm{d}q_i}{\mathrm{d}t} = \tilde{S}_i + \tilde{D}_i \tag{2}$$

will not ensure mass conservation unless special a posteriori mass fixers are introduced. When ρ_i is the prognostic variable one has to solve also an equation for the full dry mass of the atmosphere, and then evaluate the mixing ratio as $q_i = \rho_i/\rho_\mathrm{d}$ before calling chemistry.

Obtaining a formal mass conservation represented in an Eulerian grid cell from Eq. (1) requires a mass conserving Eulerian or semi-Lagrangian scheme. Traditionally, mass conservation has been obtained via Eulerian type flux based schemes (examples

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in Machenhauer et al., 2009). In recent years, a number of inherently mass conserving semi-Lagrangian schemes have been introduced: e.g. CISL (Rančić, 1992; Nair and Machenhauer, 2002), SLICE (Zerroukat et al., 2004, 2007), LMCSL (Kaas, 2008), and CSLAM (Lauritzen et al., 2010).

5.1.2 Shape preservation – monotonicity, positive definiteness

First-order accurate finite volume methods are generally attractive in the sense that they are positive definite and generally monotonic. However, since they are excessively damping for small Courant numbers, they are useless in practical applications.

Finite-volume schemes based on higher order polynomial unfiltered sub-cell representations do not, in general, fulfil requirements such as positive definiteness and monotonicity. In particular, numerical oscillations often develop near discontinuities or a large variability in gradients. It is possible to introduce different filters or constraints on the sub-grid-cell representations to reduce or eliminate these problems. Often the applications of such filters tend to reduce the accuracy of the schemes because of the implied clippings and smoothings of the sub-grid-cell polynomials. For flux-based finite volume schemes, it is also possible to introduce a posteriori corrections of the fluxes – often referred to as flux limiters – to ensure fulfilment of the desired properties (details in Machenhauer et al., 2009).

5.1.3 Wind-mass consistency

Wind-mass consistency (Byun, 1999; Jöckel et al., 2001) concerns the coupling between the continuity equation for air as a whole and for individual tracer constituents. In the non-discretized case (omitting sources, sinks and diffusion), the flux-form Eq. (3) for a constituent with mixing ratio ratio q

$$\frac{\partial q \rho_{\mathrm{d}}}{\partial t} = -\nabla \cdot q \rho_{\mathrm{d}} \boldsymbol{V} \tag{3}$$

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degenerates to

$$\frac{\partial \rho_d}{\partial t} = -\nabla \cdot \rho_d \boldsymbol{V} \tag{4}$$

for $q = 1$. This should ideally be the case numerically as well. If the two equations are solved using exactly the same numerical method, on the same grid and using the same time step, the consistency is guaranteed.

5.1.4 Spurious numerical mixing/unmixing

Recently, it has been pointed out by Lauritzen and Thuburn (2011) that since the mixing ratio of many chemical tracers are functionally related, it is highly desirable that transport schemes used in atmospheric modelling respect such functional relations and do not disrupt them in unrealistic ways. The implications of not maintaining functional relationships will generally lead to the introduction of artificial chemical reactions.

Turbulent mixing occurs in the real atmosphere. However, for chemical models operating on a fixed Eulerian grid, the mixing introduced by the numerical truncation errors is generally much stronger than the physical turbulent mixing (e.g., Thuburn and Tan, 1997). An even more severe problem, unmixing, i.e. unphysical up-gradient transport, also appears to some extent in Eulerian-based transport schemes. Lauritzen and Thuburn (2011) proposed a set of numerical tests to investigate whether a numerical transport scheme introduces unmixing or overshooting. It is strongly recommended that the numerical schemes (including all filters/limiters) used in European models – and models elsewhere – are investigated via these tests.

5.1.5 Numerical schemes implemented in European applied models

Some characteristics of online applied European models are briefly presented here. The BOLCHEM model solves the transport of passive tracers with the Weighted Average Flux (WAF) scheme (Toro, 1992) except for hydrometers to which a semi-

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Lagrangian treatment is applied (Mircea et al., 2008). The WRF-Chem (ARW dynamical core, Skamarock et al., 2005) solves fully compressible prognostic equations cast in conservative flux form for conserved variables. The transport scheme exactly conserves mass and scalar mass (Grell et al., 2005). In order to maintain consistency, monotonicity, positive definiteness and mass conservation, Enviro-HIRLAM model includes the Locally Mass Conserving Semi-Lagrangian LMCSL-LL and LMCSL-3D schemes (Kaas, 2008; Sørensen, 2012). Additionally, the model contains several options for the advection scheme previously implemented (Central Difference, Semi-Lagrangian, Bott), which does not maintain consistency between meteorology and chemistry. The usage of one or the other scheme is experiment dependent. In the NMMB/BSC-CTM, a fast Eulerian conservative and positive-definite scheme was developed for model tracers. Conservative monotonization is applied in order to control oversteepening within the conservative and positive-definite tracer advection scheme (Janjic et al., 2011).

5.2 Techniques for coupling/integration of meteorology and chemistry/aerosols

Online models are characterized by the implementation of all the chemistry processes within the meteorological driver. The meteorological information is available at each time-step directly or through a coupler. Baklanov and Korsholm (2008) define the first approach as online integrated models and the second as online access models. In the online integrated approach, two-way interactions or feedbacks are allowed between meteorology and chemistry and represent the more complete integration of AQ within meteorological processes. Computational requirements within online integrated and online access models may vary strongly. More efficient use of the computational time can be achieved with the integrated approaches, where no interpolation or double transport of passive species is performed. On the other hand, offline models are based on several independently working and to be developed components or modules (meteorology, emissions, chemistry) that exchange information through a specific interface or coupler. The main characteristic is that the information follows a one-way direction.

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For example, the results of the meteorology and the emission modules are provided to the CTM at specific times to solve the transport and fate of pollutants.

Grell and Baklanov (2011) pointed out the main strengths and disadvantages of both approaches. Online modelling systems represent the atmospheric physico-chemical processes more realistically, since the chemistry and meteorology are solved with the same time-steps and spatial grids. Thus, no interpolation in time or space is required and the same numerical schemes can be used for the transport of pollutants and passive meteorological variables. In this sense, feedback mechanisms can be considered and the model is suited for studies of aerosol effects. The inclusion of the chemistry and feedback processes may in the future improve the medium-range forecasts (2 to 5 days). On the other hand, offline modelling systems require lower computational resources. Usually, the meteorological output is already available from previous forecast or analysis runs. This allows the application only of the CTM, and provides more flexibility in specifying ensembles or performing several simulations with different inputs (e.g., different emission scenarios). This approach is probably most significant for regulatory agencies, but also for emergency response, where a multitude of ensembles can quickly be run. Grell and Baklanov (2011) pointed out that errors introduced with offline approaches will usually increase as the horizontal resolution is increased to cloud resolving scales, requiring that meteorological fields are available with much higher frequency (possibly order of minutes). For simulations on cloud resolving scales Grell et al. (2004) found significant errors in the offline approach when comparing to the online approach using the same model (MCCM).

The coupling in online models varies in complexity. Zhang (2008) identified different degrees of coupling within online models from slightly-coupled to moderately-, or fully-coupled. Not all coupled models enable a full range of feedbacks among components and processes. Selected species and processes are coupled in the slightly-coupled models, while most of the processes remain decoupled. In these systems, only specific feedbacks among processes are accounted for. In the fully-coupled models, major processes are coupled and a wide range of feedbacks are allowed. Zhang (2008) pointed

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out that few moderately- or fully-coupled online models exist in the US. This is true also for European models where only few systems account for a full range of feedbacks (Sect. 2), but there is a clear trend towards fully-coupled systems.

Examples of online integrated European models with a full-coupling approach are Enviro-HIRLAM, RAMS/ICLAMS, WRF-Chem, AQUM, and COSMO-ART. Other models do not consider all the feedbacks between meteorology and chemistry, but still maintain consistency among transport of meteorology and chemical species (e.g., BOLCHEM, MCCM, M-SYS, NMMB/BSC-CTM). As online accessible models, LOTOS-EUROS, WRF-CMAQ and MEMO/MARS couple meteorology and chemistry through an in-house or community-based coupler.

Nesting techniques allow to model domains at high horizontal resolution from information of parent grids. In online models, most efforts have been directed to the implementation of one-way nesting approaches (e.g., AQUM, BOLCHEM, COSMO-ART, M-SYS, NMMB/BSC-CTM), though some models allow also the two-way approach (e.g., WRF-Chem, MCCM, MesoNH). The consistency between nests should be carefully maintained in online models whereby feedbacks with the meteorology are turned-on. The nesting techniques implemented in online models are inherited from the mesoscale meteorological models. It is worth noting the computational impact that a nest run may have on the total wall-clock time of an execution. Computational demands rapidly increase when dealing with online models.

5.3 Computational requirements of online models and system optimizations

When developing an online coupled model, there are several computational considerations to take into account. All the traditional "good habits" should of course be applied, i.e. proper commenting, naming conventions, consistency and so on. However, from the more technical aspect one should also consider the basic structure of the code. When using online coupled models the number of prognostic variables in the model increases dramatically. To make sure that the code is still efficient, the numerical schemes must be highly multi-tracer efficient (Lauritzen et. al., 2010). All variables that can be re-used

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should be so, since the possible computing time increase for storing those variables is not negligible when using a large number of prognostic variables. The amount of data communication between the individual nodes also increases dramatically, which means that one should ensure that the communication is highly optimized, otherwise the scalability of the code can be severely limited. Since the increase in our computing power is mainly due to an increased number of processors and not clock frequency, we are faced with the problem of scalability. The models are mostly parallelized in the horizontal, meaning that, at a given resolution, we will reach a limit where it is not possible to split the domain into smaller pieces. Ideally it could be one grid box (column) per processor, however, communication, halo zones and numerical methods often limit this. For an operational model the wall-clock time becomes a hard constraint, and with the use of highly complex chemical schemes, this becomes a real problem, even if the model itself is very efficient and parallelized. It is therefore essential to choose efficient numerical schemes, which are usable in an operational setup.

The current state of online coupled models in Europe is that they are run on traditional supercomputers and written in more or less modern Fortran, which are using either Message Passing Interface (MPI), Open Multi-Processing (OpenMP), or a combination of the two for the parallelization of the code. Both methods have advantages and disadvantages, but by combining the two methods, one can ideally optimize the code for use on all types of machine architectures. In practice though, many models are only optimized for one of the methods, since writing the code for both is more cumbersome and time consuming, especially in a code under constant development. Another option is Coarray Fortran (CAF), which has not been used extensively since it was until recently only available using the Cray Fortran compiler. It was defined by Numrich and Reid in 1998 (Reid, 2010a). In 2010, it was introduced into the Fortran 2008 standard (Reid, 2010b), and is now becoming available for other compilers and machines. CAF can be implemented on shared and distributed memory computers alike, and should in either case be as fast as the OpenMP or MPI counterpart. Another benefit of using CAF is the simplification in the programming, since it is a rather sim-

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ple extension to the standard Fortran syntax. It does, however, come with a caveat: rewriting a big part of the parallelization code. Graphical Processing Units (GPUs) are often mentioned as a new possibility to achieve higher performance in our models. At this point none of the European (if any) online coupled models use GPUs alone or in combination with CPUs (Central Computing Unit). All attempts have, however, been suggesting that a very large performance gain could possibly be achieved, but this will again require rewriting large parts of the code.

5.4 Initialization and boundary values

Different approaches can be used to obtain the initial- and boundary conditions for online coupled models (both online integrated and online accessible). The methodology to prepare initial and boundary conditions does not present large differences from the procedures applied for offline models. In addition to the meteorological fields that have to be provided from the MetM in the offline approach, the 3-D distribution of chemical species have also to be known right at the beginning of the forecast, when online coupled models are used. In addition, chemical boundary values have to be specified for the length of the run.

Concerning initial fields of chemical species (chemical initial conditions), these values can either be obtained from a previous forecast using the same modelling system, global chemical initial fields from a global modelling system (such as in the MACC system), prescribed fields describing clean or polluted background atmospheres (e.g., climatological averages), or either of those methods modified with increments from a chemical data assimilation system. In offline models the improvement of the initial pollutant fields brings only a limited improvement in the forecast, because the forcing from meteorology and emissions make the model quickly converge from any reasonable initial condition to a stable solution. Indeed, a spin-up of 24–48 h is usually performed for the chemistry in such systems. As the online approach may consider interactions between meteorology and the pollutants distribution, the best possible knowledge of chemical initial conditions is required to obtain a reasonable feedback onto the mete-

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orological forecast. In this sense, the chemical initialization between online and offline models may substantially differ.

The lateral boundary values of the chemistry have to be provided at every forecast step. Since detailed information about the vertical profiles of all the chemical species are not always available, models commonly use idealized climatological profiles for boundary values, if measurement data are not available. This is especially convenient if the modelling domain is sufficiently large so that the influence of the concentrations at the boundary is small at the area of interest. With the improvement of global chemistry systems, it is becoming more common to use chemical fields from coarse model runs in the same way as for limited area meteorological forecasts.

An important problem appears when coarse or global models have different chemistry, the species have to be reordered and relumped. The following examples provide details on how the generation of boundary and initial conditions is treated in some practical applications. These difficulties can be even larger when different aerosol schemes are used, where assumptions need to be made to map from different size aerosol modes (e.g., number of size bins used to represent mineral dust) with incompatible descriptions of the aerosols composition (e.g., differences between organic matter and organic carbon mass).

Bangert et al. (2011) analyzed regional scale effects of aerosol–cloud interactions using the coupled mesoscale atmosphere and chemistry model COSMO-ART. The meteorological initial and boundary conditions are obtained from the IFS model of the ECMWF. Concerning the chemical initial fields, clean air conditions are prescribed for the gaseous and the aerosol variables. The boundary values are updated every six hours. The gaseous and the particulate species are treated at the lateral boundaries in the same way as the atmospheric variables are treated. Vogel et al. (2009) use the same approach to investigate the radiative impact of aerosol on the state of the atmosphere.

For Enviro-HIRLAM, the meteorological initial and boundary values can be obtained either from a global model or from a limited area model running a larger domain. In

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Korsholm (2009) the initial profiles for NO_x and HNO_3 are taken from the NALROM chemistry model (McKeen et al., 2002, following the procedure implemented also available in WRF-Chem), for clean Northern Hemisphere mid-latitude conditions, while the profiles for O_3 and SO_2 are measurement-based and taken from the New England Air Quality Experiment (Kleinman, 2007). For the rest of the species, climatological values along with a latitudinal, land-use and time dependent formulation from Gross (2005) are used. Aerosol number and mass concentration in nucleation and accumulation modes are climatological values based on Seinfeld and Pandis (1998). In general, the inflow of pollutants into Enviro-HIRLAM can be generated by an outer nesting and treated as the meteorological fields during the boundary preprocessing. In the boundary zone, the fields are relaxed towards the imposed fields, which include clean or dirty background values. In this way, it is possible to run nesting scenarios, whereby downscaling over a certain area is performed.

Solomos et al. (2011) used the RAMS/ICLAMS to study the effects of mineral dust and sea-salt particles on clouds and precipitation. For the meteorological initial and boundary conditions, a high resolution reanalysis dataset was used. This dataset has been prepared with the Local Analysis and Prediction System (LAPS: Albers, 1995; Albers et al., 1996), which uses an effective analysis scheme to harmonize data of different temporal and spatial resolutions on a regular grid. Two scenarios were used to define the initial values for the aerosols (clean and polluted air). The model has implemented two-way interactive nesting capabilities, which allow the use of regional scale domains together with several high resolution nested domains so that the simultaneous description of long-range transport phenomena and aerosol–cloud interactions at cloud resolving scales are possible.

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5.5 Data assimilation in online-coupled models

5.5.1 Overview of chemical data assimilation in atmospheric models

Data assimilation has been conducted for a long time in meteorological modelling and is now performed routinely for weather forecasting (Kalnay, 2003). Data assimilation in air quality modelling is more recent and although chemical data assimilation (CDA) research has progressed significantly over the past several years, operational applications are still limited. Briefly, CDA aims to improve model performance by modifying one or several components of the model: initial conditions (IC), model inputs such as emissions and boundary values or some model parameters.

Various mathematical techniques are used to assimilate the chemical observations and modify the model components. These techniques have been reviewed in literature (e.g., Carmichael et al., 2008; Zhang et al., 2012b) and can be grouped into two main categories: (1) sequential methods, which assimilate data as the model simulation proceeds forward in time (i.e., correcting the field of the state variable at successive time steps by means of a corrective term that is function of the difference between the model and the observation), and (2) variational methods, which assimilate data over a time period (4DVar) or at given time (3DVar), typically via a least-square minimization of the difference between the model and the observations. Some variational methods may require developing the adjoint of the model, which may be challenging, particularly when dealing with large chemical kinetic mechanisms and multiphase aerosol modules. Examples of sequential methods used in CDA include optimal interpolation (OI), ensemble Kalman filter (EnKF), and reduced-rank square root Kalman filter (RRSQRT).

The modification of the chemical IC is a natural extension of data assimilation as it is used in meteorology, where the chaotic nature of the primitive equations makes the system very sensitive to its initial conditions. However, modifying the chemical IC field in an AQ model typically has an effect that lasts only a few hours, because the AQ system is characterized by exponential-decay type terms reflecting atmospheric dispersion (e.g., Gaussian-type dispersion terms), chemical reactions (e.g., first- or

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second-order kinetic decay terms), and physical removal (e.g., first-order decay terms representing precipitation scavenging and dry deposition). Therefore, the atmospheric concentrations of air pollutants tend to be governed by their input functions rather than by their IC. These input functions are primarily the emission fields of primary pollutants and the boundary values. Some influential model parameters (e.g., vertical transport parameterization, kinetic rate constants) may also have a significant effect on air pollutant concentrations and are, therefore, potential candidates for improvement via data assimilation. Thus, the chemical transport component of an integrated model differs significantly from the meteorological model and, consequently, requires a different approach for data assimilation.

Recent efforts for CDA have mostly focused on CTMs, and to date only limited work has been conducted for integrated models. We briefly discuss here the observational data used in CDA, past examples of CDA in AQ modelling and, in particular, ongoing work with integrated models, and future prospects for CDA in integrated air quality modelling.

5.5.2 Observational data

The data used in CDA may come from a variety of sources. Surface air quality data are the most commonly used in AQ forecasting because they are typically readily available to the organization conducting the forecast. However, those data only provide information near the surface at a limited number of locations. Ground-based or aloft remote sensing (e.g., lidars, sondes, satellite observations) provide a more complete representation of the atmosphere albeit with other limitations: uncertainties in converting a radiance signal to a concentration, better spatial coverage but reduced temporal coverage, autocorrelation between observations, limited number of substances monitored. Clearly, there should be some advantage in using the maximum amount of information available in an optimal manner during CDA. However, the operational implementation of a CDA system that embodies a large number of data sources involves some methodological and technical challenges.

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5.5.3 Past examples of CDA in air quality modelling

Most examples of past CDA in AQ modelling concern the correction of IC of major air pollutants of interest such as O_3, NO_2 and PM. CDA has been implemented using satellite data (e.g., Elbern et al., 1997; Jeuken et al., 1999; Collins et al., 2001; Generoso et al., 2007; Boisgontier et al., 2008; Niu et al., 2008; Wang et al., 2011), ground-based concentrations (e.g., Elbern and Schmidt, 2001; Carmichael et al., 2008; Wu et al., 2008), radiosonde measurements (e.g., Elbern and Schmidt, 2001) and airborne measurements (e.g., Chai et al., 2006). For such CDA, both sequential methods and variational methods have been used.

There are also some examples of CDA for correcting emission fields, boundary values and model parameters. These studies involved inverse modelling and used variational methods. One can mention the modifications of boundary values by Roustan and Bocquet (2006), emission rates by Elbern et al. (2007) and Barbu et al. (2009), chemical reaction kinetic parameters by Barbu et al. (2009) and meteorological parameters by Bocquet (2011).

5.5.4 Current efforts on CDA in integrated models

Although most work on CDA for AQ forecasting has been conducted with CTMs, there are a few examples of recent efforts aimed at conducting CDA with integrated models. ECMWF uses the 4DVAR data assimilation developed for data assimilation in NWP to assimilate observations of atmospheric composition. In its current configuration, ECMWF's IFS (Table 4) has been extended to simulate transport, source and sink processes of atmospheric chemical species as follows (Hollingsworth et al., 2008): aerosol processes are simulated online in IFS (Morcrette et al., 2009), whereas source and sink processes of reactive gaseous species are treated via a two-way coupled global chemical transport model (Flemming et al., 2009). This coupled system has been run with MOZART-3 and TM5. This current configuration of the IFS is an intermediate step and more complex chemical kinetic mechanisms are being implemented online in IFS.

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The direct online configuration will be computationally far more efficient than the current coupled system. No interaction between atmospheric chemical composition and NWP is considered in current applications; however, research has been initiated to explore the impact of the feedbacks for instance with respect to aerosol radiative forcing (J.-J. Morcrette, 2012, personal communication) or the benefit of online CO_2 for the assimilation of satellite data (Engelen and Bauer, 2011). The use of the NWP system for data assimilation allows the use of the existing infra-structure for satellite data handling, and the MACC system is able to assimilate more than one data set from a large array of satellite instruments (GOME, MIPAS, MLS, OMI, SBUV, SCIAMACHY, MOPITT, IASI, TANSO, AIRS) for O_3, CO, NO_2, SO_2, HCHO, CH_4, CO_2 and aerosol optical depth (AOD). In the USA, CDA is conducted in WRF-Chem using both 3DVAR (Pagowski et al., 2010; Liu et al., 2011; Schwartz et al., 2012) and EnKF (Pagowski and Grell, 2012); there is an ongoing project to assimilate surface $PM_{2.5}$ data as well as AOD using a hybrid approach that employs both EnKF and 3DVAR. Furthermore, the adjoint of WRF-Chem is currently under development with the objective of performing sensitivity analysis with a variational method in the near future and possibly CDA with inverse modelling of parameter fields later. WRF-Chem is used in Europe (Table 4) and advances implemented in the USA will benefit European applications. Other models listed in Table 4 have not been used to date with CDA.

5.5.5 Recommendations for operational developments in integrated models

The significance of CDA in AQ models remains a key question. Nevertheless, it is probably the easiest CDA technique to implement because it has been widely used in meteorology and is now also used in AQ forecasting. However, one may expect more gain from CDA when used to perform inverse modelling of the emission fields. Since air quality is mostly driven by emissions, their modifications will have a longer influence than the IC. Therefore, advances in that area are likely to lead to significant benefits for integrated models. The use of CDA for parameter estimation via inverse modelling is of interest from a research point of view but its implementation for AQ

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forecasting will be more challenging than CDA of IC. Nevertheless, as a diagnostic tool, it can lead to interesting results. An additional attractiveness of the online approach in AQ modelling is, in addition to CDA, its possible usefulness for meteorological data assimilation. For example, the improved retrieval of satellite data and direct assimilation of radiances may in turn improve the forecasting of aerosol concentrations and some radiation-absorbing gases as well as day-to-day weather forecasts. Finally, it seems plausible that the knowledge of the locations of tracers plumes may – through data assimilation – improve wind fields (similar to derived wind fields from satellite cloud observations).

6 Case studies and evaluation of online coupled models

Online coupled mesoscale meteorology and chemistry models have seen a rapid evolution in the past few years particularly in the United States (see review by Zhang, 2008) and are becoming increasingly popular in Europe. Along with the increasing interest for and spread of these models, the need for a thorough evaluation through comparison with observations is growing.

Sections 6.1 and 6.2 provide an overview of application studies of online coupled meteorology and chemistry models in Europe published during approximately the past ten years. Note that in most of these studies, the coupling was only made from meteorology on chemistry, while feedbacks of chemical parameters onto the meteorology were not considered. Studies including such feedbacks are particularly highlighted in Sect. 6.2. In Sect. 6.3 the focus is on model evaluation and in particular on methodological aspects specific for online coupled models.

6.1 Applications without feedbacks

The first attempts towards online coupled atmospheric modelling in Europe considered only the transport of chemical species but not their chemical transformation (Baklanov,

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1988; Schlünzen, 1988; Kapitza and Eppel, 2000). One of the earliest studies of the full coupled chemical and meteorological evolution was the application of a coupled model during the VOTALP campaign (Vertical Ozone Transports in the ALPs) in August 1996 in southern Switzerland (Grell et al., 2000). In this study, the non-hydrostatic mesoscale model MM5 was augmented with transport of scalars and extended with modules for the simulation of chemically active species including the computation of photolysis rates, chemical reactions, biogenic emissions, and deposition. The coupled numerical model was named Multiscale Climate Chemistry Model (MCCM) and later MM5-CHEM. The simulations, which were performed in three nests at resolutions down to 1 km, depicted the complex daily thermally induced valley and mountain wind systems and demonstrated the importance of these systems for air pollutant budgets of Alpine valleys. MCCM has later been applied in various air quality studies for Europe and Mexico City, the first online coupled regional climate chemistry simulation for Europe (Forkel and Knoche, 2006), and the simulation of the 2010 Eyjafjallajökull ash plume (Emeis et al., 2011). MCCM was also used to compare online versus offline simulations on cloud resolving scales (Grell et al., 2005) to demonstrate the deficiencies of the offline approach on high resolutions.

One year before MCCM, the French mesoscale simulation system MesoNH-C for online coupling between dynamics and chemistry was introduced and applied to a pollution episode in July 1996 in the northern half of France (Tulet et al., 1999). For performance reasons, the simulations were carried out with a strongly reduced chemistry scheme but satisfactorily depicted the location and spatial extent of the pollution plume of Paris and elevated ozone levels downwind of the city. The model system was described in more detail in Tulet et al. (2003) and compared with ozone observations in France for a simulation period in August 1997. It was extended with the sectional aerosol model ORISAM (Cousin et al., 2004) as well as with the three-moments aerosol scheme ORILAM for the simulation of aerosol dynamics and secondary inorganic and organic aerosols (Tulet et al., 2005), which laid the foundation for studies of chemistry–meteorology feedbacks. MesoNH-C was subsequently employed for a wide range of

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applications to study the effect of biogenic emissions on regional ozone levels (Solmon et al., 2004), the impact of convection on aerosol hygroscopicity (Crumeyrolle et al., 2008), for regional scale CO_2 source inversion (Lauvaux et al., 2009), sulphur transport and chemical conversion in a volcanic plume (Tulet and Villeneuve, 2011), or to investigate Saharan dust transport (Bou Karam et al., 2010) to name but a few.

In the same year as MesoNH-C, von Salzen and Schlünzen (1999a,b) presented the model system METRAS coupled with gas-phase chemistry and in an online-integrated fashion with the Sectional Multicomponent Aerosol Model (SEMA). They applied the model to study the dynamics and composition of coastal aerosol in northern Germany and demonstrated the importance of sea salt aerosols for the partitioning of nitrates into the coarse mode (von Salzen and Schlünzen, 1999c). Sea salt emissions and dry deposition were calculated in direct dependence on the meteorological parameters. The model is now specifically applied for studies of pollen emission and dispersion (e.g. Buschbom et al., 2012).

Using the Regional Atmospheric Modeling Systems (RAMS) extended with online-coupled chemistry, Arteta et al. (2006) studied the impact of two different lumped chemical mechanisms on air quality simulations. Simulations were performed for the ESCOMPTE experiment conducted over Marseilles in Southern France and showed that both chemical mechanisms produced very similar results for the main pollutants (NO_x and O_3) in 3-D despite large discrepancies in 0-D (box) modelling. To judge the quality of simulations using the two schemes, the results were compared with NO_x and O_3 measurements at 75 surface stations.

The potential benefits of online coupling with respect to the quality of simulated transport and dispersion of chemical species was demonstrated by Korsholm et al. (2009). They employed the online-coupled model Enviro-HIRLAM, which can also be run offline, to study differences in the dispersion of a plume in the presence of meso-scale disturbances between online and offline representations of transport. The dispersion simulated by the online model was evaluated against data from the European Tracer

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Experiment ETEX-1 and showed satisfactory results, particularly at stations further away from the tracer release.

The Bologna limited area model for meteorology and chemistry (BOLCHEM: Mircea et al., 2008) is currently the only online coupled model operated in the EU MACC project for operational chemical-weather forecasting on the regional scale. The model recently participated in a coordinated modelling exercise to study the evolution of air pollution over Western Europe during the last decade (Colette et al., 2011).

Several groups in Europe are beginning to implement and apply the online-coupled model WRF-Chem developed primarily in the United States as a successor of MM5-Chem (see Sect. 2). Early examples are the studies by Schürmann et al. (2009) investigating the influence of synoptic and local scale meteorology and emissions on ozone concentrations in southern Italy during four selected 5–7 days periods in all seasons, and by Zabkar et al. (2011) investigating three high ozone episodes in the north-eastern Mediterranean Basin. Both studies made use of the nesting capabilities of WRF-Chem and extensively compared model simulated ozone with in-situ observations.

6.2 Applications with feedbacks

The first European model studies investigating feedbacks between chemistry and meteorology were published around 2005, firstly focusing on direct effects and later including aerosol–cloud interactions.

The impact of dust aerosols on weather forecasting in north-western Africa in September 2000 was investigated by Grini et al. (2006) using the MesoNH-C model system and for the first time considering feedbacks through the direct effect of aerosols on SW and LW radiation. They found that over the ocean dust aerosols decreased convection and over land increased vertical stability and reduced surface latent heat fluxes leading to reduced convection as well, notably over the Sahel region. They concluded that the vertical aerosol profile and single scattering albedo are particularly critical parameters and recommended that direct aerosol effects should be included in weather prediction in the Sahel region. In a similar study using MesoNH-C but ad-

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dressing a region in south-western Germany and eastern France during the Convective and Orographically-induced Precipitation Study experiment COPS, Chaboureau et al. (2011) studied the effect of Saharan dust transport to Europe on precipitation forecasts. They concluded that precipitation was better predicted when including the dust prognostic scheme and radiative feedbacks in the model.

The impact of dust aerosol radiative effects on weather forecasts was also analyzed by Pérez et al. (2006). They applied the NCEP/Eta NWP model with the DREAM model of Nickovic et al. (2001) for mineral dust transport extended with radiative feedbacks of dust aerosols on radiation to simulate a major Saharan dust outbreak over the Mediterranean in April 2002. They found significant improvements of the atmospheric temperature and mean sea-level pressure forecasts over dust-affected areas by considerably reducing warm and cold temperature biases existing in the model without dust radiation interactions. Figure 4 shows a comparison of vertical temperature profiles with radiosonde observations over the Mediterranean region most affected by the dust.

Aerosol direct effects were also studied by Vogel et al. (2009) using their new online-coupled model system COSMO-ART by comparing model simulations for two episodes in August 2005 over western Europe with and without including aerosol radiative effects. They found an average reduction of global radiation by $-6\,\mathrm{W\,m^{-2}}$ and decreases in 2 m temperatures and in temperature differences between day and night of the order of 0.1 °C each.

The two-way coupled meteorological and chemical transport modelling system MEMO/MARS-aero was used for calculating the direct aerosol effect on mesoscale meteorological and dispersion fields over the urban area of Paris, France (Halmer et al., 2010). The impact of the direct aerosol effect was found to be substantial with regard to the turbulence characteristics of the flow near the surface. High aerosol concentrations near the surface, such as those present in and around densely populated urban areas were also found to increase stability and, unlike effects at larger scales, also lead to small increases in 2 m temperatures. However, the performance of the coupled model

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in predicting urban meteorology and air quality in the specific case was only improved marginally.

European studies of aerosol indirect effects or combined direct and indirect effects are still comparatively sparse. Korsholm (2009) implemented a parameterized version of the first and second aerosol indirect effects in the Enviro-HIRLAM model system by making cloud droplet number concentrations depend on aerosol number concentrations and the autoconversion of cloud to rain droplets depend on effective cloud droplet radius. He then studied the impact of aerosol indirect effects on surface temperatures and air pollutant concentrations for a 24 h simulation over a domain in northern France including Paris in a convective case with low precipitation. He found a marginally improved agreement with observed 2 m temperatures and a marked redistribution of NO_2 in the domain, primarily as a result of the second indirect effect.

WRF-Chem has been used in various studies to investigate the impact of the aerosol interaction with radiation and microphysics outside Europe (e.g. Grell et al., 2011; Gustafson et al., 2007; Chapman et al., 2009; Zhang et al., 2010a,b, 2012c,d; Saide et al., 2012). Two European studies investigating not only the impact of aerosol direct and indirect effects on meteorology but also on air quality (O_3 and PM_{10}) were recently presented by Forkel et al. (2012) and Zhang et al. (2013). Forkel et al. (2012) applied the WRF-Chem model to simulate the two-month period June–July 2006 without any feedbacks (BASE), with aerosol direct effects only, and with both direct and indirect effects (RFBC). As shown in Fig. 5, differences in July monthly mean concentrations between the simulations RFBC and BASE had a pronounced spatial pattern and show differences in the range of 0–5 ppb for ozone and 0–5 µg m^{-3} for PM_{10} dry mass. These differences are the result of a complex interplay between small changes in surface radiative heating due to the aerosols, important semi-direct effects modifying vertical stability and cloud cover, and indirect aerosol effects which, for example, led to a substantial reduction in cloud cover over the Atlantic and hence stronger photochemical depletion of ozone over this area. Increases in PM_{10} over the Atlantic were a result of increased wind speeds in simulation RFBC as compared to BASE and

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therefore higher sea salt emissions. Over continental Europe, ABL heights were mostly reduced in simulation RFBC leading to higher PM_{10} surface concentrations particularly over the eastern part of the domain. In Zhang et al. (2013), WRF-Chem-MADRID was applied to simulate AQ in July 2001 at horizontal grid resolutions of 0.5° and 0.125° over western Europe. They found that aerosol led to reduced net SW radiation fluxes, 2 m temperature, 10 m wind speed, ABL height, and precipitation in most areas, with domain-average values of $-3.5\,W\,m^{-2}$, $-0.02\,^{\circ}C$, $-0.004\,m\,s^{-1}$, $-4.0\,m$, $-0.04\,mm\,day^{-1}$, respectively. It increases AOD and CCN over the whole domain and COT and CDNC over most of the domain.

Solomos et al. (2011) addressed the effects of pollution on the development of precipitation in both clean and polluted hazy environments in the Eastern Mediterranean by using the RAMS/ICLAMS. The model was run for a case study during 26–29 January 2009 over the Eastern Mediterranean and both direct and indirect effects were investigated, the latter not only considering the effects of aerosols as CCN but also the effect of freshly emitted mineral dust as ice nuclei. As seen in Fig. 6, the simulations showed that the onset of precipitation in hazy clouds is delayed compared to pristine conditions. Increasing the concentration of hygroscopic dust particles by 15 % resulted in more vigorous convection and more intense updrafts. Therefore, more dust particles were uplifted to higher cloud layers and acted as IN. Prognostic treatment of the aerosol concentrations in the explicit cloud droplet nucleation scheme improved the model performance for the twenty-four hour accumulated precipitation. However, the spatial distribution and the amounts of precipitation were found to vary greatly between different aerosol scenarios pointing towards large remaining uncertainties and the need for a more accurate description of aerosol feedbacks.

Aerosol indirect effects were also recently studied using the model COSMO-ART by Bangert et al. (2011) and Bangert et al. (2012). For this purpose, the model was run with the two-moment cloud microphysics scheme of Seifert and Beheng (2001) to account for the interaction of aerosols with cloud microphysics. In the first study, Bangert et al. (2011) applied the model over Europe to a cloudy five-day period in August 2005

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to study the effect of aerosols on warm cloud properties and precipitation. They found that the mean cloud droplet number concentration and droplet diameter were closely linked to changes in the aerosol. In a further study, Bangert et al. (2012) focused on the effect of mineral dust aerosols to act as ice nuclei studying an episode of Saharan dust transport to central Europe. They found the largest impact of dust on clouds at temperatures where heterogeneous freezing is dominating, thus at temperatures between the freezing level and the level of homogeneous ice nucleation. Ice crystal number concentrations were increased twofold in this temperature range during the dust event, which had a significant impact on cloud optical properties and causing a reduction in SW radiation at the surface by up to $-75\,\mathrm{W\,m^{-2}}$. The dust layer also directly caused a reduction in SW radiation at the surface which entailed a reduction in surface temperatures in the order of -0.2 to $-0.5\,^{\circ}\mathrm{C}$ in most regions affected by the dust plume and up to $-1\,^{\circ}\mathrm{C}$ in a region where regular numerical weather forecasted temperatures had been biased high by roughly the same amount.

A new advanced model system for the study of aerosol direct and indirect (liquid and ice cloud) effects based on the COSMO model and the aerosol module M7 (Vignati et al., 2004) was recently presented by Zubler et al. (2011a). The model, which is mainly aiming at climatic time scales, was subsequently applied to study the dimming and brightening in Europe due to the changes in anthropogenic aerosol loads from 1958 to 2001. Although a clear brightening from 1973 to 1998 was found in surface SW radiation under clear-sky conditions of the order of $+3.4\,\mathrm{W\,decade^{-1}}$ which is consistent with observations, no significant differences in all-sky surface SW radiation were found between the two simulations with transient and climatological (constant) emissions, as these were dominated by the evolution of cloud cover which was similar in both cases.

Although a growing number of studies applying online coupled chemistry and meteorology models have become available recently, the scope of these studies has been rather limited. For both aerosols direct and indirect effects there has been a strong focus on Saharan dust events due to their strong and readily measurable impacts on

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radiation and due to their potential role as ice nuclei. Several of these studies suggested a clear positive impact of considering aerosol feedbacks on short-term weather forecasts under such high dust-load conditions. However, more work is needed to investigate similar effects for other more commonly present aerosols of anthropogenic and biogenic origin as well as for biomass burning aerosols. More studies are also needed to address the question of whether considering feedbacks can benefit air quality forecasts both regarding summer and winter smog episodes.

6.3 Model evaluation

The evaluation of integrated meteorology-atmospheric chemistry models is a very complex task, but contributes to establishing a model's credibility by systematically assessing the model's strengths and limitations and providing guidance for the improvement of the modelling systems. A critical assessment of model performance is also important to build confidence in the use of models for research, forecasting and policy-making.

The general aims of any model evaluation are to assess the suitability of a model for a specific application ("fit for purpose"), benchmarking model performance against reality and other models, quantifying uncertainties, testing individual model components, and providing guidance for future model development. Depending on the specific aim, different evaluation strategies are required. The question of how models should best be evaluated has been addressed in numerous previous projects and publications and, therefore, only the main points are presented here.

Model evaluation is often recognized as a process of comparing model output against observations. However, although this is an important element, model evaluation may be understood in a more general sense to include all elements supporting the assessment of the quality of a model and its fitness for purpose. As suggested in a joint report of the COST Action 728 (Enhancing Mesoscale Meteorological Modelling Capabilities for Air Pollution and Dispersion Applications) and GURME (GAW Urban Research Meteorology and Environment Project) (Schlünzen and Sokhi, 2008), model evaluation should comprise the following elements

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1. *General evaluation*: Model documentation (e.g., technical report, user's guide) must exist; model must be documented in peer-reviewed literature; source-code must be open for inspection.

2. *Scientific evaluation:* Identify processes required in the model and based on these requirements evaluate suitability of model equations, approximations, parameterizations, boundary conditions, input data, etc.

3. *Benchmark tests:* Benchmarking of model performance against observations for well-defined test cases (domain and time period, model resolution, fixed input data sets including emissions and boundary conditions) and a set of quality indicators. Similarly, model performance should be analyzed for specific sensitivity tests.

4. *Operational evaluation:* This type of evaluation is specific for models used in an operational environment for example in air quality forecasting and involves operational online checking of model output, plausibility checks and quality control. However, the defined checks can also be applied in non-operational applications.

While steps 1 to 3 should be performed by a model developer and summarized in an evaluation protocol, the operational evaluation may also be performed by model users.

A less general framework for evaluating regional-scale photochemical modelling systems but more specific with respect to model benchmarking was proposed by the US Environmental Protection Agency EPA (Dennis et al., 2010) building on the concept originally proposed earlier, e.g. by Seigneur et al. (2000). It distinguishes between operational, diagnostic, dynamic (also referred to as mechanistic) and probabilistic types of model evaluation:

– *Operational evaluation* involves the direct comparison of model output with routine observations of ambient pollutant concentrations and meteorological parameters.

– *Diagnostic evaluation* examines individual processes and input drivers that may affect model performance and generally requires detailed atmospheric measurements that are not routinely available.

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– *Dynamic evaluation* investigates the model's ability to predict changes in air quality in response to changes in either source emissions or meteorological conditions. Note that with online coupled models it will also be necessary to evaluate responses in meteorology and regional climate.

– *Probabilistic evaluation* explores the uncertainty of model predictions and is used to provide a credible range of predicted values rather than a single estimate. It is based on knowledge of uncertainty embedded in both observations and model predictions, the latter often being approximated by an ensemble of model simulations.

This framework covers only steps 3 and 4 of the COST 728/GURME scheme but extends on it by separating model benchmarking into four distinctly different approaches.

Operational evaluation has been the most widely used approach in the past for both offline (Trukenmüller et al., 2004; Schlünzen and Meyer, 2007; Appel et al., 2008; Wang et al., 2009; Zhang et al., 2009a; Liu et al., 2010) and online coupled models (Zhang et al., 2010a,b, 2012c,d, 2013; Knote et al., 2011; Tuccella et al., 2012). Within the Air Quality Model Evaluation International Initiative (AQMEII), more than 20 research groups from Europe and North America recently conducted a comprehensive model evaluation exercise, the idea being to put at work the four model evaluation modes identified by Dennis et al. (2010). The aim of this exercise was to collect almost all regional-scale air quality models used for research and policy support in Europe and North America from public and private sectors and have them simulate AQ over North America and Europe for the year 2006. A large number of research and operational monitoring networks in the two continents provided a massive amount of experimental data for evaluation. This includes for the two continents one full year (2006) of continuous monitoring from almost 4000 stations for 6 gas phase species (O_3, CO, SO_2, HNO_3 and NO_2), 2700 stations of PMs, 4300 surface meteorology monitoring points, 1300 meteorological profiles at 30 locations, 800 ozonesonde profiles, and 2000+ aircraft profiles from MOZAIC. The large variety of sources of information led to a substan-

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tial effort in data harmonization and screening. All data were transferred to the JRC-ENSEMBLE system where model data were also collected as explained later (Fig. 7).

The focus was initially on operational evaluation (Solazzo et al., 2012a,b) in the sense of Dennis et al. (2010) as a first overall model assessment against the measurements. Other evaluation modes were also considered (Galmarini et al., 2012). The studies of Vautard et al. (2012), Schere et al. (2012) and Wolke et al. (2012), for example, investigated the influence of different drivers including meteorological input, grid resolution, and initial and boundary conditions, thus contributing to the diagnostic evaluation of the models. Also the study of Forkel et al. (2012) presented in Sect. 6.2 can be classified as diagnostic evaluation but different from the other studies as it addressed the question of how different processes rather than different input drivers affect the results. Solazzo et al. (2012a) conducted a multi-model ensemble analysis which is listed by Dennis et al. (2010) as a probabilistic evaluation. The activity clearly demonstrated the usefulness of such multi-model activities, the necessity of collecting harmonized monitoring information for both meteorology and chemistry, and the necessity of evaluating models in a global sense in three dimensional space and time, and in the meteorological and chemical variable space. Too often models are only evaluated against a subset of variables for a number of reasons which may lead to false conclusions since compensating mechanisms may improve one variable at the expense of others. The AQMEII exercise also revealed that a large amount of monitoring information is available but concealed or not easily accessible or simply not usable because it is not harmonized or documented.

Dynamic evaluation has been applied in numerous regional modeling studies relating observed changes in ozone and/or aerosol concentrations to anthropogenic emission changes (e.g., Jonson et al., 2006; Zhang et al., 2009b, 2013; van Loon et al., 2007; Vautard et al., 2007; Gilliland et al., 2008; Godowitch et al., 2011; Colette et al., 2011; Hogrefe et al., 2011; Zubler et al., 2011b). A considerable uncertainty in these regional trend studies turned out to be the influence of lateral boundary conditions – stressing

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the importance of suitably embedding a regional model into a larger scale or global model.

In the context of regional online coupled models, interactions between meteorology and chemistry through aerosol direct and indirect effects are of particular interest. Evaluating the representation of these processes requires a comprehensive assessment of the various processes influencing aerosol distributions, their physical and chemical properties, and consequently their effects on radiation and cloud and rain formation (see Sects. 2 and 4 for an overview of processes and interactions). An important difference from evaluations of offline CTMs is that the evaluation of meteorological parameters is equally important as the evaluation of trace gas and aerosol parameters.

Most evaluation studies of online coupled regional models performed so far (e.g., Zhang et al., 2010a,b, 2012c,d, 2013; Knote et al., 2011) followed approaches that had been successfully applied to offline models for many years as exemplified by phase 1 of AQMEII, but these are not sufficient to emphasize the specific advantages of online modelling. Previous assessments of model representations of chemistry–meteorology feedbacks were mainly restricted to comparing simulations with and without the respective interaction. Demonstrating that adding feedbacks improves model performance when compared with observations, however, has been and will remain a great challenge and will require new and improved strategies for model evaluation. Assessing feedbacks typically involves comparing small differences between simulations with and without a given feedback mechanism and evaluation of differences is inherently more challenging than evaluation of absolute levels. In many cases, judging whether including a given feedback improves model performance or not will only be possible by integrating over a sufficiently long time (or a sufficient number of simulations) to distinguish signal from noise and will require mature models that are sufficiently close to reality and not employ much data simulation to allow for model internal feedbacks. Both requirements pose great challenges since long-term simulations are computationally expensive and the details of many feedback mechanisms, particularly of aerosol–cloud feedbacks, are only poorly understood and can only be represented in a highly parame-

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terized way in the models. It also places high demands on observational data sets since many parameters required for a thorough evaluation are not routinely measured, for example, photolysis rate of NO_2, size resolved chemical aerosol composition, aerosol size distributions and optical properties, SW and LW radiation fluxes, AOD, CCN and IN activity, cloud droplet number concentrations and size distributions. Future measurement campaigns should therefore be planned carefully in collaboration with modellers to meet the needs of assessing chemistry–meteorology feedbacks in online integrated regional models.

The improvements of forecasts resulting from online coupled models with feedbacks need to be critically addressed by the community in a well-coordinated way as is currently planned under the auspices of AQMEII phase 2 in collaboration with the European COST Action EuMetChem. Detailed lists of chemistry–meteorology interactions to be considered in model evaluation studies and observational datasets available for model evaluation, as recommended by EuMetChem and AQMEII, are given in Tables B1–B3.

7 Conclusions and recommendations

The aim of this section is to highlight selected scientific issues and emerging challenges that require proper consideration to improve reliability and usability of the online integrated meteorology-chemistry modelling approach for three main communities: CWF and AQ, NWP, and climate.

In this paper, we have reviewed the current status of online air quality and meteorology modelling illustrated with 18 models developed or applied in Europe. All the selected models can address regional scale phenomena with horizontal resolutions in the range 1–20 km, and applications ranging from the global scale (e.g., IFS-MOZART, Met-UM, NMMB/BSC-CTM, WRF-Chem) to urban scale (e.g., Enviro-HIRLAM, MCCM, GEM-AQ, Meso-NH, M-SYS, NNMB/BSC-CTM, RAMS/ICLAMS, WRF-Chem, WRF-CMAQ), down to the local scale (M-SYS, WRF-Chem). All the models are applicable

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to study episodes and some of them also to long-term runs and climate studies (e.g., MEMO/MARS, RACMO2/LOTOS-EUROS, WRF-Chem, WRF-CMAQ) or regional climate studies (e.g., BOLCHEM, COSMO-ART, MCCM, M-SYS, RACMO2/LOTOS-EUROS, WRF-Chem). Feedbacks of pollutants on meteorology are already considered in most of these models (COSMO-ART, COSMO-MUSCAT, Enviro-HIRLAM, MCCM, MEMO/MARS, Meso-NH, MetUM, NMMB/BSC-CTM, RACMO2/LOTOS-EUROS, RAMS/ICLAMS, RegCM-Chem4, REMO-HAM/REMOTE, WRF-Chem, WRF-CMAQ). However, the models differ considerably in the number of interactions and the level of details of the process representations. Besides, not all of them realise the online integration of meteorology and chemistry, some models (e.g. RACMO2/LOTOS-EUROS, COSMO-MUSCAT) follow the online access approach, with a data exchange between the chemistry and meteorology modules not on each model time step.

The great challenge for the community of online coupled modellers is to ensure that the incorporation of these complex and computationally-demanding feedback mechanisms improves the final results and can contribute to the ensemble of reliable models in Europe.

7.1 Major challenges and needs

7.1.1 Integrating European research

The COST Action ES1004 EuMetChem was launched in 2011 with the aim to develop a European framework for online integrated air quality and meteorology modelling. The Action aims to foster the exchange of knowledge, to review the current state-of-the-art in online coupled modelling, and to make recommendations on processes (considering their relevance for different applications), on best coding practices, model evaluation strategies, and applications. Rather than developing a single model, the Action seeks to provide recommendations for efficient interfacing and integration of modules in order to facilitate the exchange of codes developed by different research groups. A better coor-

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dination and integration of European efforts is clearly needed to address the numerous challenges within the field of atmospheric chemistry–meteorology modelling.

7.1.2 Interacting processes and feedback mechanisms

The focus on integrated systems is timely, 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.

An expert poll was conducted amongst the members of the COST Action and other experts from different countries around the world to identify the most important meteorology and chemistry interactions in online MetChem models. The results (summarized in Table 3) show that the perceived most important interactions differ from one application category to another. In general, most of the meteorology and chemistry interactions are more important for CWF models than for NWP and climate models, and those interactions are represented better in CWF models, according to this expert opinion poll. Primary attention needs to be given to interactions that have been ranked as "high" in importance and, at the same time, as "not represented well" in models, such as the improvement of "aerosol indirect effects" (for NWP/Climate) and "effect of liquid water on wet scavenging and atmospheric composition" as well as "wind speed – dust/sea-salt interactions" (for CWF models).

The processes particularly critical for online coupling between the chemical and meteorological components include: *advection, convection* and *vertical diffusion* (control the transport and dispersion of chemical species and hence critically affect surface concentrations); *cloud microphysics* (determines cloud life cycle, interacts with aerosols, and affects soluble chemical species); *radiative transfer* (fluxes determined by meteo-

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rological parameters and radiatively active chemical compounds); and *drag interactions* (influence on wind and further on the distribution of chemical species).

The description of the ABL structure and of a range of processes including radiative transfer, cloud microphysics and precipitation formation should be improved. Convection and condensation schemes need to be adjusted to take the aerosol–microphysical interactions into account, and the radiation scheme needs to be modified to include accurately the aerosol effects. Hence, it is recommended to build a library of different available parameterisations of aerosol forcing mechanisms, aerosol–cloud interactions, radiation schemes, etc.

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 affects gas phase chemistry but also the coupling with aqueous phase and aerosol mechanisms, have faced practical difficulty 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. All modelling groups should be encouraged to resubmit their own mechanisms, even if they only revise slightly an existing mechanism.

- 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.

The interaction of aerosols with gas-phase chemistry and their impacts on radiation and cloud microphysics depend strongly on their physical and chemical properties.

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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.

Furthermore, aerosols have a strong impact on cloud processes such as cloud microphysics and physical properties as well as on precipitation release. Therefore, all online coupled models have cloud schemes that to some extent represent the effect of aerosols on clouds. However, 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 (especially for ice forming nuclei, interaction with cirrus clouds, contribution of different anthropogenic and biogenic/natural aerosol particles for cloud evolution, etc.). On the other hand, the inclusion of aerosol effects in convective parameterizations is only beginning to get attention.

Online coupling imposes additional requirements on the setup and implementation of radiation parameterizations. Most of these requirements reflect the need to maintain physical and numerical consistency 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 parameterisation needs to be is currently not possible.

Finally, emissions and deposition also interact in a specific way with the meteorological core model within online-coupled models. The most interesting emissions are those which depend on meteorology as these could potentially be treated more accurately and consistently than in offline models. Natural emissions closely 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

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online. Emissions of biogenic volatile organic compounds (BVOC) like isoprene and terpenes and of pollen are also strong functions of meteorological conditions and calculated correspondingly in models.

7.1.3 Numerical and computational aspects

A general overview on numerical and computational aspects of online European models was presented in Sect. 5. Current state and desired characteristics of the models were addressed in terms of numerical methods, coupling techniques, computational aspects, boundary conditions and chemistry data assimilation methods.

The desirable numerical properties of transport schemes have been outlined. The most relevant properties to be considered when developing integrated models, and especially for CTM modelling, 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 European integrated models.

A detailed analysis of the numerical properties of European integrated models is recommended. A particularly relevant set of tests has recently 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 acts like introducing artificial chemical reactions in the system.

The techniques of model coupling have been addressed throughout the paper. A clear trend towards fully integrated model development is becoming perceptible in Europe with five model systems that can be considered as fully integrated models with all relevant feedbacks implemented. Complementing those, there are several ones that are built using an integrated approach, but some major feedbacks are not included yet. A third group of models, the online-access models, is characterized by applying an external coupler between meteorology and chemistry. All the information is passed

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through the coupler. Depending on the approach used, consistency problems may arise. In this sense, fully integrated models are desirable for a better representation of feedback processes.

Numerical performance is also an important issue. The current parallelization is based on well established MPI and OpenMP programming models. Beyond these approaches there is no clear trend towards new parallelization 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.

7.1.4 Data assimilation

Experience with chemical data assimilation (CDA) in integrated models is still limited. First efforts have been engaged with a couple of integrated systems (IFS-MOZART and WRF-Chem). The easiest approach is probably the adjustment of initial conditions (IC) through CDA. In a similar manner to meteorological data assimilation, optimal interpolation, variational approaches or EnKF are applicable. Other methodologies such as inverse modelling of emission fields appear as a promising technique to improve the skill of integrated models, and may have a stronger impact for short-lived pollutants than data assimilation for IC. 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.

7.1.5 Evaluation of methodologies and data

There is a crucial need for an 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 ap-

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plied to online integrated modelling as well. The NWP community has the necessary tools, for example, to analyze whether including certain feedbacks or not has a positive effect on weather forecast skills. 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 (not widely available), chemical composition and optical properties are needed. Such observations should ideally be collocated with detailed radiation measurements (e.g., 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 scarcely 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. Again, long simulations are needed to demonstrate the benefits of online coupling in a statistically significant way. Due to the importance of aerosols for chemistry–meteorology interactions, more long-term observations of aerosol composition and size distributions and of the aerosol precursors such as HNO_3, NH_3 and VOCs would be desirable.

7.2 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, NWP, AQ/CWF, and climate modelling. However, there is not necessarily one integrated online modelling approach/system suitable for all communities.

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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 time steps and grid cells. This also includes harmonized parameterizations 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 non-linearities involved, offline coupling can lead to inaccuracies in chemical composition simulations.

For NWP modelling, the advantages of online approaches are less evident and need to be further investigated and justified. It is clear that online models for 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 and very simplified chemistry (with a focus on aerosol precursors and formation, e.g., sulphur chemistry).

For climate modelling, the feedbacks (forcing mechanisms) are the most important and the main improvements are related to climate–chemistry/aerosols interactions.

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However, the online approach is not critical for all purposes. Many GCMs or RCMs are using an offline approach for describing greenhouse gas (GHG) and aerosol forcing processes (by chemistry/aerosol parameterization 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 (Folberth et al., 2011) that for long-lived GHG forcing the online approach did not give large improvements. On the other hand, for short-lived climate forcers (SLCF), 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 SLCF, which represent one of the main uncertainties in current climate models and are in particular at the core of political and socio-economical assessments of future climate change mitigation strategies. It will be impossible to answer the main questions about SLCFs and mitigation strategies without fully online integrated modelling systems that include aerosol dynamics and feedbacks.

Based on the analysis of the models included in this paper we suggest that we should aim to eventually migrate from separate MetM and CTM systems to fully integrated online 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, where, e.g., water vapour and other atmospheric gases are no longer treated numerically differently only because they came traditionally from different disciplines. The integration has also the advantage of saving 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, and costs for both types of models.

The main recommendations from this study are briefly summarised in the following list. If a recommendation is mainly relevant for one type of the application (**Met**eorology or **Chem**istry simulations) this is explicitly mentioned:

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7.2.1 Emissions

- Meteorology-dependent emissions (e.g., biogenic, sea spray, windblown dust, lightning, pollen) need to be parameterized more accurately and improved for Chem in online integrated or coupled Met-Chem models.
- Emissions from ships and aviation as well as emissions and heat fluxes from forest fires, volcanic eruptions, etc., need to be better known and improved in the models for both application types.
- Anthropogenic VOC emissions need to be improved for Chem simulations.
- Emissions of primary aerosols and their number emissions need to be better represented for Chem simulations.
- Ammonia fluxes should be bi-directional, which is mainly relevant for Chem simulations.

7.2.2 Model formulations

- Eventually migrating from offline to online integrated modelling systems 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. While this may be the obvious approach for Chem simulations, more research and discussion may be needed for Met simulations. CWF and NWP communities should work more closely together.
- The online access modelling approach is less effective (and more expensive) than the online integration of meteorology with atmospheric chemistry and aerosol dynamics and thus should be aimed at.

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- Our parameterisation/understanding of aerosol–radiation–cloud–chemistry interactions is still incomplete and further research on the model representations of these interactions is needed.

- Aerosol properties (number, aerosol mass, composition, size distribution, phase, hygroscopicity, mixing state, and optical depths) and processes (chemistry, thermodynamics, and dynamics) need to be better represented for Chem simulations, in particular, several areas of high uncertainty such as the formation of SOA should be focused upon.

- Cloud properties (droplet number concentrations, size distribution, cloud fraction, liquid water content, optical depths), processes (microphysics, dynamics, in-cloud and below-cloud 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, which could be tested and evaluated against laboratory/field studies would greatly facilitate this transition.

- Data assimilation in online models can include both meteorological and chemical data assimilation. However, as more variables are assimilated into a model, one must be cautious about possible diminishing returns as well as possible antagonistic effects due to the interactions between meteorological variables and chemical concentrations. Consequently, future work is warranted in this area to develop optimal methods for data assimilation in online meteorological/air quality models.

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7.2.3 Real-time application

- National weather centres should consider progressively including aerosol/chemistry interactions into NWP systems and extending their forecasts to chemical weather as well. Chemical species and their processes may help identifying shortcomings in the transport schemes of NWP models and may lead to improved assimilation of meteorological satellite data through a better representation of the effect of gases and aerosols on radiative transfer.
- Centres responsible for CWF should seriously consider online modelling as a necessary part of their suite of forecasts. Additional advantages will arise from cross evaluations for both disciplines.
- The frequency of integration between meteorology and chemistry models needs to be large enough to at least properly consider the effects of meso-scale events in high-resolution CTMs. This will yield ways to online integrated approach.
- For CWF, online coupling of meteorology, physics, and emissions and their accurate representations are very important; implementation of aerosol feedbacks is important mostly for specific episodes and extreme cases.

7.2.4 Model evaluation

- An international testbed for model evaluation for urban- and meso-scale would be very useful and is really needed for integrated Met-Chem models. A first step into this direction has been taken by the AQMEII consortium for the regional scale.
- Special variables (e.g., shortwave and longwave radiation, photolytic rate of NO_2, AOD, COT, CCN, cloud droplet number concentration, precipitation) should be included for a systematic model evaluation for online-coupled models, in addition to those used for offline-coupled models. This, however, also means that reliable measurements need to be provided on a routine base.

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– To verify meteorology/climate–chemistry feedbacks, additional measurements that are not available in routine ground-based networks such as radiative forcing, PBL height, photolytic rates of NO_2 should be included. Group-based and satellite remote-sensing measurements and retrievals of aerosol and cloud properties (e.g., optical depths, radiative forcing, CCN, IN, cloud droplet number concentrations) are very useful to validate such feedbacks. Long term measurement data of meteorological variables such radiation fluxes, aerosol and cloud properties, and aerosol mass and number concentrations are urgently needed.

Appendix A

Model descriptions

Short model descriptions are given here. Tabular overviews can be found at http://www.mi.uni-hamburg.de/costmodinv.

A1 BOLCHEM, Italy

BOLCHEM (BOLAM + CHEM; Mircea et al., 2008) is a model based on a project that started in 2002 at CNR-ISAC. Gas and aerosol transport, diffusion, removal and transformation processes are included in the existing (and continuously developed and maintained) BOLAM meteorological hydrostatic limited area model (Buzzi et al., 2003). Applications range from regional to hot-spot, with time scales ranging from air quality forecasts to climatological ones. Equations for atmospheric dynamics, thermodynamics, radiation, microphysics and chemistry are solved simultaneously on the same spatio-temporal grid making every feedback potentially available.

BOLCHEM ran in forecast mode over Europe during the GEMS project (http://gems.ecmwf.int/; Huijnen et al., 2010; Zyryanov et al., 2012) and is currently running to forecast Air Quality over Italy in the MACC project (http://www.gmes-atmosphere.eu/). It

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also participated within the CityZen project (http://www.cityzen-project.eu) in regional trend and future scenario studies (Colette et al., 2011). In addition, several specific studies were performed: volcanic emission event (Villani et al., 2006), forest fire episodes (Pizzigalli, 2012), aerosol direct effects (Russo et al., 2010), Saharan dust transport over the Mediterranean Sea (Mircea et al., 2008), composition data assimilation (Messina et al., 2011), scale bridging technique (Maurizi et al., 2012).

A2 COSMO-ART, Germany

COSMO-ART is a regional to continental scale model (ART stands for Aerosols and Reactive Trace gases, Vogel et al., 2009), online-coupled to the COSMO regional numerical weather prediction and climate model (Baldauf et al., 2011). COSMO is used by several European countries for operational weather forecast. The gaseous chemistry in COSMO-ART is solved by a modified version of the Regional Acid Deposition Model, Version 2 (RADM2) mechanism (Stockwell et al., 1990), which is extended to describe secondary organic aerosol formation and hydroxyl recycling due to isoprene chemistry and heterogeneous reactions as hydrolysis of N_2O_5. Aerosols are represented by the modal aerosol module MADEsoot (Riemer et al., 2003). The five modes that represent the aerosol population contain: pure soot, secondary mixtures of sulphates, nitrates, ammonium, organics and water (nucleation and accumulation) and the internal mixtures of all these species in both modes. Separate fine and coarse emission modes for sea-salt, dust (Stanelle et al., 2008), and rest anthropogenic species are treated by six additional modes. Specific modules are included to simulate the dispersion of pollen grains (Vogel et al., 2008) and other biological particles. Meteorology-affected emissions are also online-coupled to the model system. The equilibrium between phases of the inorganic material is achieved through the ISORROPIA II module (Fountoukis and Nenes, 2007). The simulation of secondary organic aerosol chemistry and of organic mass transfer between phases in COSMO-ART is currently treated with the SORGAM scheme (Schell et al., 2001). That scheme was recently replaced by a VBS (volatility basic set) scheme (Athanasopoulo et al., 2013). The radiation scheme used within the

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model to calculate the vertical profiles of SW and LW radiative fluxes is GRAALS (Ritter and Geleyn, 1992). Radiative fluxes are online modified by the aerosol mass, and its soot fraction. In order to account for the interaction of aerosol particles with the cloud microphysics and radiation COSMO-ART uses the two moment cloud microphysics scheme of Seifert and Beheng (2006) and parameterizations of cloud condensation and ice nuclei (Bangert et al., 2011, 2012). A first evaluation of the model system can be found in Knote et al. (2011).

A3 COSMO-MUSCAT, Germany

The multiscale model system COSMO-MUSCAT (Wolke et al., 2004a,b, 2012) is qualified for process studies as well as the operational forecast of pollutants in local and regional areas. Different horizontal resolutions can be used for individual sub-domains in the developed multi-block approach, which allows finer grid sizes in selected regions of interest (e.g., urban areas or around large point sources). The operational forecast model COSMO (Steppeler et al., 2003) of the German Meteorological Service (DWD) is online coupled with the chemistry transport model MUSCAT, which treats the atmospheric transport as well as chemical transformations for several gas phase species and particulate matters. The transport processes include advection, turbulent diffusion, sedimentation as well as dry and wet deposition. The chemical reaction system RACM-MIM2 (Karl et al., 2006; Stockwell et al., 1997) with 87 species and over 200 reactions is applied successfully in 3-D air quality applications and case studies. For the description of the particle size distribution and aerosol dynamical processes the modal aerosol model M7 (Vignati et al., 2004) has been extended by nitrate and ammonium. In this approach, the total particle population is aggregated from seven log-normal modes with different compositions. The gas-to-particle partitioning of inorganic species is performed using the thermodynamic aerosol model ISORROPIA (Nenes et al., 1998). Alternatively, a more simplified mass based particle model is available especially for long-term simulations.

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The modelling system has been used for several air quality applications (Stern et al., 2008; Hinneburg et al., 2009; Renner and Wolke, 2010) and the investigation of the large-scale transport of Saharan dust, including its sources and sinks (e.g., Heinold et al., 2007; Helmert et al., 2007). In addition to parameterizing particle fluxes and transformations, the influence of aerosols by modifying solar and thermal radiative fluxes on temperature, wind fields, and cloud dynamics are considered (Heinold et al., 2011a; Meier et al., 2012). Furthermore, the distribution of the Volcano ash plume over Europe has been analyzed (Heinold et al., 2011b).

A4 Enviro-HIRLAM, Denmark and HIRLAM countries

Enviro-HIRLAM is developed as a fully online coupled NWP and Chemical Transport model for research and forecasting of joint meteorological, chemical and biological weather. The integrated modelling system is developed by DMI and other collaborators (Chenevez et al., 2004; Baklanov et al., 2008a, 2011b; Korsholm et al., 2008, 2009; Korsholm, 2009) and included by the HIRLAM consortium as the baseline system in the HIRLAM Chemical Branch; it is used in several countries. The model development was initiated at DMI more than 10 yr ago. The first version of Enviro-HIRLAM was based on the DMI-HIRLAM NWP model with online integrated pollutant transport and dispersion (Chenevez et al., 2004), chemistry, deposition and indirect effects (Korsholm, 2009) and aerosol dynamics (Baklanov, 2003; Gross and Baklanov, 2004). To make the model suitable for chemical weather forecasting in urban areas the meteorological part was improved by implementation of urban sub-layer parameterizations (Baklanov et al., 2008b). The model's dynamic core was improved by adding a locally mass conserving semi-Lagrangian numerical advection scheme (Kaas, 2008; Sørensen, 2012), which improves forecast accuracy and makes it possible to perform longer runs. The current version of Enviro-HIRLAM (Nuterman et al., 2013) is based on reference HIRLAM version 7.2 with a more sophisticated and effective chemistry scheme, multi-compound modal approach aerosol dynamics modules, aerosol feedbacks on radiation (direct and

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semi-direct effects) and on cloud microphysics (first and second indirect effects). This version is still under development and needs further validation.

The modelling system is used for operational pollen forecasting in Denmark since 2009 and for different research studies since 2004. Following the main development strategy of the HIRLAM community (HIRLAM-B project), the Enviro-HIRLAM further developments will be moving towards the new HARMONIE NWP platform by incorporation of the Enviro-HIRLAM chemistry modules and aerosol–radiation–cloud interactions into the future Enviro-HARMONIE integrated system (Baklanov, 2008; Baklanov et al., 2011a).

A5 GEM-AQ, Canada (used in Poland)

GEM-AQ model (Kaminski et al., 2008) is a comprehensive chemical weather model in which air quality processes (chemistry and aerosols) and the tropospheric chemistry are solved online in the operational weather prediction model GEM. GEM is the Global Environmental Multiscale model, developed at Environment Canada (Côté, et al., 1998). Recently, the model was extended to account for chemistry-radiation feedback, where modelled (chemically active) ozone, water vapour and aerosols are used to calculate heating rates. For regional Arctic simulations the model chemistry was extended to account for reactive bromine species in order to investigate ozone depletion in the boundary layer (Toyota et al., 2011).

The GEM-AQ model in LAM configuration is used in a semi-operational air quality forecast for Europe and Poland (e.g., Struzewska and Kaminski, 2008). The model is run on several regional domains with horizontal resolutions of ∼ 15 km (whole Europe), ∼ 5 km (Poland) and ∼ 1 km for agglomerations (Krakow), where urban effects are represented by the TEB (Town Energy Balance) parameterization (Masson, 2000).

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A6 IFS-MOZART (MACC/ECMWF)

The meteorological forecast and data assimilation system IFS (Integrated Forecast System, http://www.ecmwf.int/research/ifsdocs) at the European Centre for Medium-Range Weather Forecast (ECMWF) has been coupled to an updated version of the global chemistry transport model MOZART-3 (Model for Ozone And Related Tracers, version 3; Kinnison et al., 2007) in order to build the coupled MACC system IFS-MOZART (Flemming et al., 2009). For a coupled simulation both models are running in parallel and exchange meteorological fields as well as 3-D source and sink terms every hour using the OASIS4 coupling software developed in the PRISM project (Valcke and Redler, 2006). The coupled system is currently used to provide analysis and forecast of atmospheric composition (http://www.gmes-atmosphere.eu). The coupled system will be superseded by the online coupling of the chemical mechanisms into the IFS (C-IFS) following the implementation of aerosol modules (Morcrette et al., 2009).

A7 MCCM, Germany

The online coupled regional meteorology-chemistry model MCCM (Mesoscale climate chemistry model, Grell et al., 2000) was developed at the IMK-IFU. MCCM is based on the non-hydrostatic NCAR/Penn State University mesoscale model MM5. It offers the choice between the tropospheric gas phase chemistry mechanisms RADM, RACM, and RACM-MIM. BVOC emissions and photolysis are calculated online. Aerosol is described by the modal MADE/SORGAM aerosol module. Like MM5, MCCM can be applied from the continental to the urban scale.

Applications of MCCM were various air quality studies for Europe and Mexico City, the first online coupled regional climate chemistry simulation (Forkel and Knoche, 2006), or the simulation of the 2010 Eyjafjallajökull ash plume (Emeis et al., 2011).

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A8 MEMO/MARS, Greece

The MEMO/MARS-aero modelling system combines the mesoscale meteorological model MEMO (Moussiopoulos et al., 1997) with the chemical dispersion model MARS-aero (Moussiopoulos et al., 1995) in an online or offline coupling configuration. The aerosol phase is described in MARS-aero as a multimodal (fine, accumulation and coarse) internally mixed distribution. For inorganics, an equilibrium model has been built especially for dry, coastal and urban regions, which contains common inorganic species and also crustal species. For secondary organics (SOA), the SORGAM module has been incorporated into the model. Radiative effects of air pollutants and cloud layers are introduced in the coupled configuration using an extended version of the radiation module IRIS (Halmer, 2012) which incorporates the OPAC (Optical Properties of Aerosols and Clouds) software library (d'Almeida et al., 1991). OPAC defines a dataset of typical clouds and internally mixed aerosol components, which can be externally mixed to simulate a wide range of tropospheric aerosols. The MEMO/MARS-aero modelling system forms the operational core of the Air Quality Management System used by the environmental ministry of the Republic of Cyprus (Moussiopoulos et al., 2012). The performance of the coupled system was evaluated in an urban case application for Paris, France (Halmer et al., 2010) by analysing the response of the primary meteorological variables and dispersion fields to the introduction of the direct aerosol effect.

A9 Meso-NH, France

Meso-NH is a non-hydrostatic mesoscale atmospheric model coupled online with chemistry, which has been jointly developed by CNRM (Météo France) and Laboratoire d'Aérologie (CNRS) (Lafore et al., 1998). Meso-NH simulates synoptic scale (horizontal resolution of several tens of kilometres) to small scale (LES type, horizontal resolution of a few meters) and can be run in a two-way nested mode. The model is used for research for both meteorological and chemical weather. Different sets of parameterization are included in the model for convection (Bechtold et al., 2001), for cloud micro-

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physics (Pinty and Jabouille, 1998; Cohard and Pinty, 2000; Khairoudinov and Kogan, 2000), for turbulence (Cuxart et al., 2000), for biosphere-atmosphere thermodynamic exchanges (Noilhan and Mahfouf, 1996) and for urban-atmosphere interactions (Masson, 2000). The physical package dedicated to the mesoscale has been included in the numerical weather prediction model AROME that is operational since 2008 on France at 2.5 km resolution.

For the chemistry part, the model includes online gaseous chemistry (Suhre et al., 2000; Tulet et al., 2003), online aerosols chemistry (Tulet et al., 2005) and online cloud chemistry including mixed phase cloud (Leriche et al., 2012). Several chemistry mechanisms are available for the gas phase. The RACM and the ReLACS (Crassier et al., 2000), which is a reduced mechanism from RACM, are dedicated to the modelling of ozone, NO_x and VOC chemistry system in the troposphere. The CACM (Caltech Atmospheric Chemistry Mechanism, Griffin et al., 2002) and the ReLACS2 (Tulet et al., 2006), which is a reduced mechanism from CACM, are dedicated, in addition to the modelling of the ozone, NO_x, VOC chemistry system, to the modelling of the semi-volatile organic compounds, precursors of SOA formation.

A10 MetUM (Met Office Unified Model), UK

The Met Office Unified Model (MetUM) (Davies et al., 2005) uses the aerosol scheme CLASSIC (Bellouin et al., 2011) that previously was applied in climate and air quality configurations. The chemistry scheme is the UKCA scheme (Morgenstern et al., 2009; O' Connor et al., 2013) which offers several choices of chemical mechanism. A two moment modal aerosol scheme, UKCA-GLOMAP-mode, has also been developed for use with the MetUM ported from the offline model TOMCAT (Mann et al., 2010). The model is two-way coupled with the direct radiative effects of gases/aerosols and the indirect effects of aerosols capable of being treated.

MetUM is used across a very wide range of spatial and temporal scales from short range weather forecasting at 1.5 km resolution to multi–decadal simulations in an earth system model configuration (Collins et al., 2011).

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A11 M-SYS (online version), Germany

The multi-scale model system M-SYS (Trukenmüller et al., 2004) is a community model with the development coordinated at the University of Hamburg. M-SYS combines the non-hydrostatic MEsoscale TRAnsport- and Stream model METRAS (Schlünzen, 1990; Schlünzen and Pahl, 1992; resolutions of a few km to a few hundred metres) with the obstacle-resolving MIcroscale model MITRAS (Schlünzen et al., 2003; Bohnenstengel et al., 2004; resolution a few metre) using 1-way nesting in dependence of application and characteristic scales as outlined by Schlünzen et al. (2011). Both models calculate transport and solve 3-D gas-phase chemistry (RADM2 mechanism, Stockwell et al., 1990) and aerosol reactions, when coupled to the corresponding chemistry modules (MECTM/MICTM). Advection and diffusion are solved with the same numerical schemes for the different scales and meteorology and chemistry. For resolutions of at least 1 km the sectional aerosol model SEMA is employed (von Salzen and Schlünzen, 1999a,b). Early applications, e.g. to coastal (von Salzen and Schlünzen, 1999c), biogenic emissions (Renner and Münzengern, 2003) or urban areas (Schlünzen et al., 2003) revealed that online modelling was too resources consuming at that time and hindered scientific progress. Therefore, the coupling time step was reduced to 15 min to 3 h, depending on the application (e.g. Lenz et al., 2000; Müller et al., 2001; Schlünzen and Meyer, 2007; Meyer and Schlünzen, 2011). However, increasing computer power allows reducing the coupling interval and including direct impacts, e.g. for pollen emissions which directly depend on meteorology or for pollen fertility which is radiation dependent (Schueler et al., 2006). Other processes solved in direct dependence of meteorology are, besides the transport processes and radiation changes due to clouds, deposition (Schlünzen an Pahl, 1992) and sedimentation (von Salzen and Schlünzen, 1999a) and biogenic emissions. The models consider several sub-gridscale land uses per grid cell and employ a flux aggregation approach (von Salzen et al., 1996) to more realistically describe typical surface characteristics (Schlünzen and Katzfey, 2003); this is also considered in the online coupled sea-ice model, where several ice classes plus

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water might occur in one grid cell (Lüpkes and Birnbaum, 1995). To better describe urban effects the BEP scheme has been included (Grawe et al., 2012).

Applications of the online integrated system concern atmospheric inputs into the marginal seas and mud-flat areas (Schlünzen and Pahl, 1992; Schlünzen et al., 1997; von Salzen and Schlünzen, 1999c), aerosol load and concentrations within street canyons (Schlünzen et al., 2003) or biogenic emissions and gene flow on a landscape level (Renner and Münzenberg, 2003; Schueler and Schlünzen, 2006; Buschboom et al., 2012).

A12 NMMB/BSC-CTM (BSC-CNS), Spain

The model NMMB/BSC-CTM (NMMB/BSC Chemical Transport Model) is a new fully online chemical weather prediction system under development at the Earth Sciences Department of the Barcelona Supercomputing Center in collaboration with several research institutions (National Centers for Environmental Predictions (NCEP), NASA Goddard Institute for Space Studies, University of California Irvine). The basis of the development is the NCEP new global/regional Non-hydrostatic Multiscale Model on the B grid (NMMB; Janjic et al., 2011; Janjic and Gall, 2012). Its unified non-hydrostatic dynamical core allows regional and global simulations and forecasts. A mineral dust module has been coupled within the NMMB (Pérez et al., 2011). The new system simulates the atmospheric life cycle of the eroded desert dust. The main characteristics are its online coupling of the dust scheme with the meteorological driver, the wide range of applications from meso to global scales, and the dust shortwave and long-wave radiative feedbacks on meteorology. In order to complement such development, an online gas-phase chemical mechanism has been implemented (Jorba et al., 2012). Chemical species are advected and mixed at the corresponding time steps of the meteorological tracers using the same numerical scheme of the NMMB. Advection is Eulerian, positive definite and monotone. The final objective of the work is to develop a fully chemical weather prediction system, namely NMMB/BSC-CTM, able to resolve gas–aerosol–meteorology interactions from global to local scales. Current efforts are

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oriented to incorporate a multi-component aerosol module within the system with the aim to solve the life-cycle of relevant aerosols at global scale (dust, sea salt, sulphate, black carbon and organic carbon).

A13 RACMO2/LOTOS-EUROS, the Netherlands

The regional climate model RACMO2 (Van Meijgaard et al., 2008) is online coupled to the regional chemistry transport model LOTOS-EUROS (Schaap et al., 2008). They are coupled through a 3-hourly exchange of meteorology and aerosol concentrations, and the system therefore has some features of an online access model. In addition to differences in internal time steps, both models run on their native grids (rotated pole for RACMO2 versus regular lat-lon for LOTOS-EUROS), with a typical resolution of 25 km, although a resolution of the order of 10 km is also feasible. Since LOTOS-EUROS only covers the lowest 3.5 km of the atmosphere, as it was designed as an air quality model, RACMO2 has to use climatology for the rest of the vertical. A vertical extension of LOTOS-EUROS is being developed.

RACMO2 is a semi-Lagrangian model based on the dynamics of the HIRLAM model, combined with the physics of the ECMWF IFS system. It has taken part in ensemble studies with other regional climate models and is used for the downscaling of climate scenarios for the Netherlands. LOTOS-EUROS is a Eulerian model, using CBM-IV for gaseous chemistry and EQSAM (Metzger et al., 2002) for secondary inorganic aerosols. Secondary organics are not accounted for yet. Biogenic, dust and sea spray emissions are calculated online. The model currently uses a bulk approach for aerosol ($PM_{2.5}$ and PM_{10}) although M7 (Vignati et al., 2004) is available. LOTOS-EUROS is part of the MACC ensemble and has taken part in EURODELTA and AQMEII model intercomparison exercises. It is used in the Netherlands for smog forecasting and policy-oriented studies. A one-way coupled version was used to study the impact of climate change on air quality (Manders et al., 2012). A two-way coupled version including the direct impact of aerosol on radiation (Savenije et al., 2012) and on cloud condensation number (Van Meijgaard et al., 2012) is now available.

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A14 RAMS/ICLAMS, USA/Greece

The Integrated Community Limited Area Modelling System (ICLAMS; Solomos et al., 2011) has been developed at the University of Athens, with contributions from ATMET LLc, USA and Georgia Institute of Technology, as an extended version of RAMS6.0 atmospheric model (Cotton et al., 2003). It is a new generation integrated modelling system that includes two-way interactive nesting, detailed surface (soil, vegetation) and explicit cloud microphysics. The desert dust module of SKIRON (Spyrou et al., 2010) has been implemented in RAMS/ICLAMS and the model includes also a sea-salt module, gas and aqueous phase chemistry, heterogeneous chemical processes and an improved radiation scheme (RRTM).

Photodissociation rates, radiative transfer corrections as well as aerosol–cloud interactions are calculated online. The same radiative transfer scheme is used for both physical and photochemical processes. All prognostic aerosols in the model are allowed to act as CCN/GCCN/IN for the activation of cloud droplets and ice particles following the formulations of Nenes and Seinfeld (2003) and Barahona and Nenes (2009). CCN/GCCN/IN are treated in an explicit way.

The model capabilities make RAMS/ICLAMS appropriate for studying complicated atmospheric processes related to chemical weather interactions and quantifying forcing from them.

A15 RegCM-Chem, Italy

RegCM4-Chem is the ICTP-Regional Climate Model online coupled with the atmospheric chemical transport model. The climate component of the coupled model is the RegCM. The chemistry component in RegCM4 depends on the condensed gas-phase chemistry which is based on CBM-Z (Zaveri and Peters, 1999). During the last years the RegCM has been coupled with simplified chemistry/aerosol modules of increasing complexity, such as a simplified sulphur chemistry scheme including direct and indirect aerosol radiative effects (Qian and Giorgi, 1999; Qian et al., 2001), a simple carbon

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aerosol module (Solmon et al., 2006), a desert dust model (Zakey et al., 2006) and a sea salt scheme (Zakey et al., 2008).

Studies of regional chemistry-climate interactions with the RegCM system include the effects of direct effects of sulphate on the climate of east Asia (Giorgi et al., 2002, 2003), the effects of desert dust on the African monsoon (Konare et al., 2008; Solmon et al., 2008, 2012), the effect of European aerosol (Zanis et al., 2012), and the effects of dust storms on East Asia climate (Zhang et al., 2007).

A16 REMOTE/REMO-HAM, Germany

The regional three-dimensional online climate–chemistry/aerosol model REMOTE (Regional Model with Tracer Extension) is based on the former regional weather forecast system of the German Meteorological Service (Majewski, 1991), extended by gas phase and aerosol chemistry. A basic description is available from Langmann (2000). For the determination of aerosol dynamics and thermodynamics, the M7 module is implemented (Vignati et al., 2004). The aerosol dynamical processes in M7 include nucleation, coagulation and condensation. The aerosol size spectrum is represented by the superposition of seven log-normal distributions subdivided into soluble and insoluble coarse, accumulation and Aitken modes and an additional soluble nucleation mode. The five aerosol components considered in M7 are sulfate, black carbon, organic carbon, sea salt and mineral dust (Langmann et al., 2008). Photochemical production and loss in REMOTE are determined by the RADM II chemical scheme (Stockwell et al., 1990). Based on REMOTE, the REMO-HAM has recently be developed and evaluated (Pietikäinen et al., 2012). It uses the same chemical mechanism but is based on a newer version of the meteorology model REMO (B. Langmann, 2013, personal communication).

Several evaluation studies and applications in different regions of the Earth and for different kinds of aerosols (anthropogenic emissions, mineral dust, volcanic emissions, biomass burning emissions) and feedback studies focusing on cloud–aerosol feed-

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backs have been performed (e.g. Coleman et al., 2013; Langmann et al., 2012; O'Dowd et al., 2012; Pfeffer et al., 2012).

A17 WRF-Chem, USA, used in Germany, UK, Spain and others

The Weather Research and Forecast (WRF; http://www.wrf-model.org/) model coupled with Chemistry (WRF-Chem; Grell et al., 2005; Fast et al., 2006) provides the capability to simulate chemistry and aerosols from cloud scales to regional scales. WRF-Chem is a community model. The development is lead by NOAA/ESRL with contributions from National Center for Atmospheric Research (NCAR), Pacific Northwest National Laboratory (PNNL), EPA, and university scientists (http://www.wrf-model.org/WG11). WRF-Chem is an online model which includes the treatment of the aerosol direct and indirect effect. Standard gas phase chemistry options of WRF-Chem include the RADM2 and the CBMZ mechanism, additional chemistry options are available with a preprocessing tool based on KPP. For the aerosol it offers the choice between bulk, modal, and sectional schemes. The Volatile Basis Set (VBS) approach is also available for the modal and sectional aerosol approaches to treat Secondary Organic Aerosol formation. Among other options MEGAN may be used for biogenic emissions, two pre-processors are available for wildfires (injection heights are being calculated online).

WRF-Chem is used for research applications or for forecasting of air quality (e.g. http://verde.lma.fi.upm.es/wrfchem_eu), volcanic ash dispersion, and weather. Due to its versatility WRF-Chem is attracting a large user and developer community world-wide and also in Europe. WRF-Chem is continually further developed and additional options are implemented. References from model applications and/or developments can be found at http://ruc.noaa.gov/wrf/WG11/References/WRF-Chem.references.htm. There are also several versions and branches/lines of the modelling system under development (see e.g., Zhang, 2008, 2010a, 2012c, 2013; Li et al., 2010).

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A18 WRF-CMAQ Coupled System, USA (used in UK)

The Community Multiscale Air Quality (CMAQ) modelling system (Byun and Schere, 2006) developed by USEPA since 1990s is one of the decision tools for regulatory applications. Traditionally meteorological models are not built in the CMAQ model. The users need to run a meteorological model, like the Fifth-Generation Pennsylvania State University-National Center for Atmospheric Research Model (MM5) or Weather Research and Forecasting (WRF) (Skamarock et al., 2008) model firstly, then use the meteorological model output to drive CMAQ. There is no chemistry feedback to meteorology in the offline manner.

The new version CMAQ 5.0 (Officially released February 2012, http://www.cmaq-model.org/) includes an option to run the model in a 2-way coupled (online) mode with the WRFv3.3 model (Pleim et al., 2008; Mathur et al., 2010; Wong et al., 2012). A coupler is used to link these two models, ensuring exchange between the meteorology and atmospheric chemistry modelling components. In this 2-way coupled system, simulated aerosol composition and size distribution are used to estimate the optical properties of aerosols, which are then used in the radiation calculations in WRF. CMAQv5.0 includes a new version of the SAPRC gas-phase chemical mechanism – SAPRC07TB (Carter et al., 2010), the new version CB05 with updated toluene chemistry (Whitten et al., 2010) and a new aerosol module AERO6. These to be used with CAM and RRTMG (two radiation schemes options in WRF-CMAQ) calculate the aerosol extinction, single scattering albedo, and asymmetry factor for short-wave (SW) radiation and aerosol extinction for long-wave (LW) radiation. The aerosol chemical species calculated by CMAQ are combined into five groups: water-soluble, insoluble, sea-salt, black carbon, and water. The refractive indices for these species are taken from the OPAC database (Hess et al., 1998) using linear interpolation to the central wavelength of the CAM and RRTMG wavelength intervals (Wong et al., 2012).

Offline WRF-CMAQ has been used in a number of UK and projects throughout Europe for air quality regulatory applications, such as EU FP7 MEGAPOLI,

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TRANSPHORM. Appel et al. (2012) have demonstrated the WRF-CMAQ modelling system used in the AQMEII (phase 1) model evaluation. The online 2-way coupled WRF-CMAQ model will be used in AQMEII (phase 2) within the online-coupled model evaluation exercise and to explore air quality and climate change interactions.

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Table 1. Meteorology's impacts on chemistry.

Temperature	Modulates chemical reaction and photolytic rates Modulates biogenic emissions (isoprene, terpenes, dimethyl sulphide, etc.) Photo-synthetically active radiation: modulates biogenic emissions (isoprene, monoterpenes) Affects the volatility of chemical species Controls aerosol dynamics (coagulation, condensation, nucleation)
Temperature vertical gradients	Determines vertical diffusion intensity
Temperature and humidity	Affect aerosol thermodynamics (e.g., gas/particle partitioning, secondary aerosol formation)
Water vapour	Modulates OH radicals, size of hydrophilic aerosol
Liquid water	Determines wet scavenging and atmospheric composition
Cloud processes	Affects mixing, transformation and scavenging of chemical compounds
Precipitation	Wet removal of trace gases and aerosol
Soil moisture	Modulates dust emissions Influences dry deposition (biosphere and soil)
Lightning	Increases NO_x emissions
Radiation	Modulates chemical reactions and photolytic rates Modulates biogenic emissions (isoprene, monoterpenes)
Wind speed and direction	Determines horizontal transport and vertical mixing of chemical species Influences dust emissions and sea-salt emissions
ABL height	Influences concentrations

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Table 2. Chemical species' impacts on meteorology.

Aerosols	Modulate radiation transfers (SW scattering/absorption, LW absorption) Affect boundary layer meteorology (temperature, humidity, wind speed, ABL height, stability) Extraordinary high concentrations can affect stability and wind speed Act as cloud condensation nuclei
Aerosols physical properties (size distribution, mass and number concentrations, hygroscipicity)	Influences cloud droplet or crystal number and hence cloud optical depth and hence radiation Modulates cloud morphology (e.g., reflectance) Influence precipitation (initiation, intensity) Affects haze formation and atmospheric humidity Change scattering/absorption
Soot deposited on ice	Changes albedo
Radiatively-active gases	Modulate radiation transfers

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Table 3. Summary results (based on 30 participants) of the expert survey on the most important meteorology and chemistry interactions for online MetChem models. Only the top six ranked important interactions (out of 24 total interactions in the survey questionnaire) for each model category (NWP, CWF and climate) are reported here. Score 1 ranks the "importance of the interaction" from the weighted mean of: 4 = high, 3 = medium, 2 = low and 1 = negligible; while score 2 ranks the "adequacy of the representation of the interaction in models" as the weighted mean of: 4 = quite well; 3 = fairly well; 2 = poor; 1 = not included.

Rank	Top six ranked Meteorology and chemistry interactions Changes in … affect (–>) …	Score1	Representation in models (%) Quite well	Fairly well	Poor	Not incl.	Don't know	Score2
(A) Numerical Weather Prediction (NWP)								
1	aerosol –> precipitation (initiation and intensity of precipitation)	**3.1**	0	8.3	54.2	25	12.5	**1.6**
2	aerosols –> radiation (shortwave scattering/absorption and longwave absorption)	**3.1**	8.3	20.8	45.8	16.7	8.3	**2.0**
3	temperature vertical gradients –> vertical diffusion	**3.1**	4	64	8	8	16	**2.3**
4	aerosol –> cloud droplet or crystal number density and hence cloud optical depth	**3.1**	4	8	44	28	16	**1.6**
5	aerosol –> haze (relationship between the hygroscopic growth of aerosols and humidity)	**2.9**	0	4	44	32	20	**1.3**
6	aerosol –> cloud morphology (e.g., reflectance)	**2.7**	0	8	48	32	12	**1.5**
Averaged score for all 24 interactions in NWP models		**2.3**	3.1	13.1	30.9	36.7	16.3	**1.5**
(B) Chemical Weather Forecast (CWF)								
1	wind speed –> dust emissions and seasalt emissions	**3.8**	7.7	42.3	46.2	0	3.8	**2.5**
2	precipitation (frequency/intensity) –> atmospheric composition	**3.8**	14.3	57.1	21.4	0	7.1	**2.7**
3	temperature –> chemical reaction rates and photolysis	**3.7**	32.3	54.8	0	0	12.9	**2.9**
4	radiation –> chemical reaction rates and photolysis	**3.7**	20	53.3	13.3	0	13.3	**2.7**
5	liquid water –> wet scavenging and atmospheric composition	**3.6**	11.5	23.1	57.7	0	7.7	**2.3**
6	temperature vertical gradients –> vertical diffusion	**3.6**	3.7	70.4	14.8	3.7	7.4	**2.6**
Averaged score for all 24 interactions in CWF models		**3.1**	8.3	35.3	36.5	6.2	13.7	**2.2**
(C) Climate modelling								
1	aerosols –> radiation (shortwave scattering/absorption and longwave absorption)	**3.4**	16.7	41.7	16.7	0	25	**2.3**
2	radiatively active gases (e.g., water vapour, CO_2, O_3, CH_4, NO and CFC) –> radiation	**3.4**	12	36	20	0	32	**2.0**
3	aerosol –> precipitation (initiation and intensity of precipitation)	**3.0**	0	20	40	4	36	**1.4**
4	radiation –> chemical reaction rates and photolysis	**3.0**	10.3	20.7	24.1	3.4	41.4	**1.5**
5	aerosol –> cloud droplet or crystal number density and hence cloud optical depth	**3.0**	4	12	44	0	40	**1.4**
6	temperature –> chemical reaction rates and photolysis	**3.0**	16.7	36.7	13.3	0	33.3	**2.0**
Averaged score for all 24 interactions in climate models		**2.8**	5.9	20.3	31.2	3.9	38.6	**1.5**

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Table 4. Online integrated or online access Meteorology-Chemistry models developed or applied in Europe. Typical grid-sizes are from 1 km to 20 km.

N	Model, Country, Web-site	Meteorology component	Gas phase chemistry (gpc) & aerosol module (amo) components	Feedback of pollution to meteorology DAE – Direct aerosol effect IAE – Indirect aerosol effect	Applications ER – episodes run LR – long-term runs	Scale G – global H – hemispheric C – continental R – regional U – urban L – local
1	BOLCHEM, Italy http://bolchem.isac.cnr.it	BOLAM	SAPRC90 gpc, $AERO_3$ amo	Under development	CWF, climate (ER)	C –> R
2	COSMO-ART, Germany http://www.imk-tro.kit.edu/3509.php	COSMO	Extended RADM2 gpc, modal amo, soot, pollen, mineral dust, volcanic ash	DAE on radiation, IAE	Climate mode (ER)	C –> R
3	COSMO-MUSCAT, Germany[b] http://projects.tropos.de/cosmo_muscat	COSMO	RACM gpc, 2 modal amo, mineral dust module	DAE on radiation for mineral dust	(ER)	C –> R
4	Enviro-HIRLAM, Denmark and HIRLAM countries http://hirlam.org/chemical	HIRLAM[a]	NWP and CBM-Z gpc, modal and sectional amo, liquid phase chemistry	DAE and IAE	CWF (ER)	H –> R –> U
5	GEM-AQ Canada and Poland http://ecoforecast.eu	GEM	ADOM-IIb gpc	DAE on radiation, IAE (in-cloud chemistry and aerosol formation)	ER	C –> R
6	IFS-MOZART (MACC/ ECMWF), C-IFS http://www.gmes-atmosphere.eu	IFS	MOZART gpc with updates to JPL-06, MACC amo	DAE and IAE	Forecasts, Reanalysis (ER)	G
7	MCCM, Germany http://www.imk-ifu.kit.edu/829.php	MM5	RADM, RACM or RACM-MIM2 gpc with modal amo	DAE	climate–chemistry (ER)	C –> R –> U
8	MEMO/MARS, Greece http://pandora.meng.auth.gr/mds/showlong.php?id=19	MEMO	RACM gpc, 3 modal amo, SOA based on SORGAM	DAE	(ER & LR)	R –> U
9	Meso-NH, France http://mesonh.aero.obs-mip.fr/mesonh	Meso-NH	RACM, ReLACS or CACM gpc, modal amo	DAE	(ER)	C –> R –> U –> L
10	MetUM (Met Office Unified Model), UK http://www.metoffice.gov.uk/research/modelling-systems/unified-model	MetUM	2 tropo- and 1 stratospheric chem. schemes, 2 alternative aerosol schemes	DAE and IAE, radiative impacts of N_2O, O_3, CH_4	CWF, climate-chemistry studies (ER)	G –> R
11	M-SYS (**online version**), Germany http://www.mi.uni-hamburg.de/SYSTEM-M-SYS.651.0.html	METRAS	RADM gpc, sectional amo, pollen module	None, radiative impacts of O_3, CH_4	(ER)	R –> U –> L
12	NMMB/BSC-CTM (BSC-CNS), Spain http://www.bsc.es/earth-sciences/mineral-	NMMB	BSC-mineral dust scheme CBM-IV and CBM05 chemical mechanisms	DAE on radiation for mineral dust	Forecast, Reanalysis (ER)	G –> U
13	RACMO2/LOTOS-EUROS[b] (KNMI, TNO), Netherlands http://www.knmi.nl/research/regional_climate/models/racmo.html http://www.lotos-euros.nl	RACMO2	CBM-IV and EQSAM chemistry, sectional approach ($PM_{2.5}$, PM_{10})	DAE, Effect of aerosol on CCN	Climate & policy oriented studies	R

[a] New version of the model based on the HARMONIE meteorological core is under development.

[b] The COSMO-MUSCAT and RACMO2/LOTOS-EUROS systems are not online models and only partly/conditionally can be included into the category of online access Meteorology-Chemistry models, because the MetM and CTM interfaces not on each time step, but they start implementing some feedback mechanisms.

[c] Besides the official version of WRF-Chem mentioned here, there exists several other versions, e.g., by Li et al. (2010), MARDID: Zhang et al. (2010a, 2012d, 2013).

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Table 4. Continued.

N	Model, Country, Web-site	Meteorology component	Gas phase chemistry (gpc) & aerosol module (amo) components	Feedback of pollution to meteorology DAE – Direct aerosol effect IAE – Indirect aerosol effect	Applications ER – episodes run LR – long-term runs	Scale G – global H – hemispheric C – continental R – regional U – urban L – local
14	RAMS/ICLAMS, USA/Greece http://forecast.uoa.gr/ICLAMS/index.php	RAMS	Online photolysis rates, coupled SAPRC99 gas phase, modal amo, ISORROPIA equilibrium and SOA, cloud chemistry	DAE and IAE	CWF, meteo-chemistry interactions (ER)	C –> U
15	RegCM-Chem4, Italy http://users.ictp.it/RegCNET/model.html or http://gforge.ictp.it/gf/project/regcm	RegCM4	CBM-Z, uni-modal amo, sectional mineral dust, sulphur chemistry	DAE	Climate-chemistry	C –> R
16	REMO-HAM/REMOTE, Germany http://www.remo-rcm.de/The-REMO-model.1190.0.html	REMO	RADM gas phase, Walcec&Taylor liquid phase, M7 (Vignati et al., 2004)	GHGs effects on radiation	(ER) (e.g., volcanic ash), climate	C –> R
17	WRF-Chem[c], US (**used in Germany, UK, Spain, etc.**) http://ruc.noaa.gov/wrf/WG11	WRF	RADM, RACM, RACM-MIM with modal amo (also VBS approach for Organics) or CBM-Z/CB05 with sectional amo MOSAIC and MADRID, liquid phase chemistry, SAPRC99 with MOSAIC, MOZART 4 with bulk aerosols (GOCART) or MOSAIC	DAE and IAE	CWF, climate–chemistry (ER)	C –> R, G –> U –> LES
18	WRF-CMAQ Coupled System, USA (**used in UK**) http://www.epa.gov/amad/Research/Air/twoway.html	WRF	gpc: CB05 with updated toluene chemistry, SAPRC07TB; AERO6 amo	DAE on radiation and photolysis	Episodes to annual (ER & LR)	H –> U

[a] New version of the model based on the HARMONIE meteorological core is under development.
[b] The COSMO-MUSCAT and RACMO2/LOTOS-EUROS systems are not online models and only partly/conditionally can be included into the category of online access Meteorology-Chemistry models, because the MetM and CTM interfaces not on each time step, but they start implementing some feedback mechanisms.
[c] Besides the official version of WRF-Chem mentioned here, there exists several other versions, e.g., by Li et al. (2010), MARDID: Zhang et al. (2010a, 2012d, 2013).

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Table 5. Meteorology models currently used as basis for coupled models.

NWP model	References/documentation	Continuity Eq. Approx.	Advection	Convection	Vertical diffusion	Radiation	Underlying meteorology component in CTM
BOLAM	http://www.isac.cnr.it/ dinamica/ bolam/index.html	Incompressible, hydrostatic	Weighted Average Flux (Toro, 1992)	Kain and Fritsch (1993)	Prognostic TKE	Mixed: Morcrette (1991); Ritter and Geleyn (1992)	BOLCHEM
COSMO	Baldauf et al. (2011), Steppeler et al. (2003) http://www.cosmo-model.org/content/model/documentation/core/default.htm	Non-hydrostatic	Semi-Lagrangian, Lin and Rood (1996), Bott (1993)	Moist: Tiedtke (1989). Option for the Kain–Fritsch (1993) scheme Shallow: Reduced Tiedtke scheme	Prognostic TKE	δ two-stream radiation scheme after Ritter and Geleyn (1992)	COSMO-ART, COSMO-LM-MUSCAT
ECWMF-IFS	http://www.ecmwf.int/research/ifsdocs/CY38r1/index.html	Non-hydrostatic	Semi-Lagrangian (Hortal, 2002)	Mass-flux scheme described in Bechtold et al. (2008)	Based on local Richardson number and Monin-Obukov profile (Beljaars and Viterbo, 1999)	SW: Morcrette (1991) LW: ECMWF version of the RRTM scheme (Morcrette, 1998)	C-IFS, EHAM5/6-HAMMOZ
GEM	Côté et al. (1998) http://collaboration.cmc.ec.gc.ca/science/rpn/gem	Hydrostac and Non-hydrostatic depending on resolution	Semi-Lagrangian	Kuo-type deep convection scheme Kain and Fritsch (1993)	Prognostic TKE	LW: Garand (1983), Garand and Mailhot (1990) SW: Fouquart-Bonnel Correlated K Li and Barker (2005)	On-line in the GEM model
HARMONIE	http://hirlam.org/index.php?option=com_content&view=article&id=65&Itemid=102	Compressible non-hydrostatic	Semi-Lagrangian	As AROME	As AROME	ACRANEB (Ritter and Geleyn, 1992)	Enviro-HIRLAM/ HARMONIE
HIRLAM	http://hirlam.org/index.php?option=com_content&view=article&id=64&Itemid=101	Hydrostatic and non-hydrostatic versions	Semi-Lagrangian	Modified STRACO (Sass and Yang, 2002) or Kain and Fritsch (1993)	CBR Cuxart et al. (2000)	Savijärvi (1990)	Enviro-HIRLAM
MEMO	http://pandora.meng.auth.gr/mds/showlong.php?id=19	Non-hydrostatic	TVD and FCT schemes	NA	Prognostic TKE	LW, SW: Moussiopoulos (1987), Halmer (2012)	MEMO/MARS
Meso-NH	http://mesonh.aero.obs-mip.fr/mesonh/	Non-hydrostatic	2nd order difference eulerian schemes	Mass flux (Bechtold et al., 2001)	Turbulence scheme Cuxart et al. (2000)	LW: RRTM (Mlawer et. al., 1997); SW: Fouquart (1980)	Meso-NH
METRAS	Schlunzen et al. (2012) http://www.mi.uni-hamburg.de/692.html	Anelastic, non-hydrostatic	Adams Bashfort scheme with centred or up to 3rd order (W)ENO (Schroeder et al., 2006) advection	Explicit scheme for clouds by for-atmosphere turbulence, counter gradient scheme for shallow con-vection (Lupkes and Schlunzen, 1996)	Choice of different schemes, normaly chosen: maximum of Blackadar and counter gradient scheme (Lupkes and Schlunzen, 1996)	LW and SW calculated Using 2-stream approximation	M-SYS

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Table 5. Continued.

NWP model	References/documentation	Continuity Eq. Approx.	Advection	Convection	Vertical diffusion	Radiation	Underlying meteorology component in CTM
MM5	Grell et al. (1994), NCAR Tech Note TN-398 + STR, http://www.mmm.ucar.edu/mm5/ http://www.mmm.ucar.edu/mm5/ documents/mm5-desc-doc.html	Non-hydrostatic	Leap frog with Asselin filter For tracers: Smolarciewicz and Grabowski (1990)	Choice between Anthes–Kuo, Grell, Arakawa–Schubert, Fritch–Chapell, Kain–Fritsch, and Betts–Miller-scheme	MM5: choice between Blackadar, Burk–Thomson, ETA, MRF, Gayno–Seaman, and Pleim–Chang scheme; for MCCM limited to Burk–Thomson scheme	Choice between "Cloud" (Dudhia), CCM3, and RRTM scheme	MCCM
NMMB	Janjic and Gall (2012)	Non-hydrostatic	Eulerian, Adams Bashforth (Janjic and Gall, 2012)	Betts–Miller–Janjic (Janjic, 2000)	Prognostic TKE	RRTM (Mlawer et al., 1997)	NMMB/ BSC-CTM
RACMO2	http://www.knmi.nl/research/regional_climate/ uploads/models/FinalReport_CS06.pdf	Hydrostatic	Semi-Lagrangian	Tiedtke (1989), Nordeng (1994), Neggers et al. (2009)	Lenderink and Holtslag (2004), Siebesma et al. (2007)	Fouquart and Bonnel (1980), Mlawer et al. (1997)	LOTOS-EUROS
RAMS	Cotton et al. (2003) http://www.atmet.com/	Non-hydrostatic or hydrostatic	Hybrid combination of leapfrog and forward in time (Tremback, 1987)	Modified Kuo (Tremback 1990) Kain–Fritsch cumulus parameterization	Smagorinsky (1963), Lilly (1962) and Hill (1974). Deardorff and Mellor-Yamada level 2.5 Isotropic TKE	L&SW: Chen and Cotton (1983), Harrington (1997), Solomos et al. (2011) RRTM Mlawer et al. (1997), Iacono et al. (2000)	RAMS/ ICLAMS
RegCM4	http://www.ictp.it/research/esp/models/regcm4.aspx	Hydrostatic	Weighted Average Flux Semi-lagrangian	mass-flux cumulus scheme (Grell, 1993; Tiedke, 1989)	Holtslag and Bouville (1993), UW pbl (Bretherton et al., 2004)	CCM3 Kiehl et al. (1996), RRTM/McICA, Mlawer et al. (1997)	RegCM-Chem4, EnvClimA
REMO	http://www.remo-rcm.de/The-REMO-model.1190.0.html	Hydrostatic	Second order horizontal and vertical differences	Mass-flux convection scheme after Tiedke (1989)	Louis (1979) in Prandtl layer, ext. level-2 scheme Mellor and Yamada (1974) in Ekman layer and free-flow, modif. for clouds	Delta-two-stream radiation scheme after Ritter and Geleyn (1992)	REMOTE, REMO-HAM
MetUM	Davies et al. (2005), http://www.metoffice.gov.uk/research/ modelling-systems/unified-model	Non-hydrostatic for latest version	Semi-Lagrangian	Lock et al. (2000)	Lock et al. (2000)	Edwards and Slingo (1996)	MetUM
WRF	Skamarock et al. (2008), http://www.wrf-model.org/index.php	non-hydrostatic, fully compressible	RK3 scheme, described in Wicker and Skamarock (2002)	Modified Kain and Fritsch (1993), Grell and Devenyi (2002)	Prognostic TKE	SW: Goddard; Dudhia LW: RRTM	WRF-Chem WRF-CMAQ

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Table 6. Comparison of chemical mechanisms used in coupled models. In the photolysis rate column, "+" means documentation available does not separately list the photolysis reactions and so they are included in the chemical reactions. NA means the available documentation did not include the relevant data. Note also that several of these mechanisms are explicitly gas phase chemistry mechanisms – the models in which they are implemented may include aqueous phase chemistry in addition.

Mechanism	Chem species	Chem rxns	Photol rxns	Het. rxns	Aq. chem	Model(s)	Reference(s)
ADOM-IIb	50	100	+	NA	NA	GEM	Venkatram et al. (1988)
CBM-IV (aka CB4)	33	81	+	NA	NA	NMMB/BSC-CTM, BOLCHEM, RACMO2/ LOTOS-EUROS	Gery et al. (1989)
CBM-05 (aka CB05)	52	156	+	NA	NA	NMMB/BSC-CTM, WRF-CMAQ	Sarwar et al. (2008)
CBM-Z		132	+	NA	NA	RegCM-Chem, Enviro-HIRLAM, WRF-Chem	Zaveri and Peters (1999)
GEOS-CHEM	80	> 300	+	N_2O_5 and NO_3 to nitric acid in sulphate	NA	RegCM-Chem (under testing)	Bey et al. (2001)
MECCA1	116	295	+	NA	NA	MESSy(ECHAM5)	Sander et al. (2005)
MOZART2	63	132	32	N_2O_5 and NO_3 on sulphate	NA	ECHAM5/6-HAMMOZ	Horowitz et al. (2003)
MOZART3	108	218	18	71	NA	IFS-MOZART	Kinnison et al. (2007)
MOZART4	85	157	39	4	NA	ECHAM5/6-HAMMOZ, WRF-Chem	Emmons et al. (2010)
NWP-Chem	17–28	27–32	4	NA	17	Enviro-HIRLAM v1	Korsholm et al. (2008)
RADMK	86	171	22	1	NA	COSMO-ART	Vogel et al. (2009)
RADM2	63	136	21	NA	NA	MCCM, M-SYS, REMO, WRF-Chem; M-SYS	Stockwell et al. (1990)
RACM	77	214	23	NA	NA	COSMO-LM-MUSCAT, MCCM, Meso-NH, RegCM-Chem, MEMO/ MARS, WRF-Chem	Stockwell et al. (1997)
RACM-MIM	84	221	23	NA	NA	MCCM, WRF-Chem	Geiger et al. (2003)
RAQ (plus CLASSIC)	61	115	23	NA	Oxidation of SO_2 by H_2O_2 and O_3	MetUM	Collins et al. (1997, 1999)
ReLACS	37	128	+	NA	NA	Meso-NH	Crassier et al. (2000)
SAPRC90 SOA	43	131	16	NA	NA	BOLCHEM	Carter (1990)
SAPRC99	72	198	+	NA	NA	RAMS/ICLAMS, WRF-CMAQ, WRF-Chem	Carter (2000)
SAPRC07	44–207	126–640	+	NA	NA	WRF-CMAQ	Carter (2010)
StdTrop (plus CLASSIC)	42	96	25	NA	Oxidation of SO_2 by H_2O_2 and O_3	MetUM	Law et al. (1998)

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Table 7. Approaches of aerosol physics applied in different models.

Name of model	Approach	Number of modes or bins, comments	References
BOLCHEM	Modal	3	Binkowski et al. (2003)
CHIMERE	Sectional	6	Vivanco et al. (2009)
CMAQ AERO5/AERO6	Modal	3 (aitken, accumulation and coarse)	Byun and Schere (2006)
COSMO-ART	Modal	11	Vogel et al. (2009)
Enviro-HIRLAM v1	Modal	3	Baklanov (2003), Gross and Baklanov (2004), Korsholm (2009)
GAMES	Sectional	10	Carnevale et al. (2008)
ICLAMS, Enviro-HIRLAM v2, REMO-HAM/REMOTE	Mixed, depending on process and aerosol type	M7 aerosol module (configurable)	Vignati et al. (2004), Solomos et al. (2011)
LOTOS-EUROS	Sectional	2	Schaap et al. (2008)
MetUM	Mass only	CLASSIC. 8 aerosol species, dust has 2 or 6 size bins, other aerosols bulk scheme with 2 or 3 modes each	Bellouin et al. (2011)
MetUM	Modal	UKCA-GLOMAP-mode, based on M7 approach, configurable.	Bellouin et al. (2013)
MCCM, WRF/Chem	Modal	2 (modules MADE-SORGAM, MADE-VBS)	Ackermann et al. (1998), Schell et al. (2001)
M-SYS	Sectional	4 to 64	Von Salzen and Schlünzen (1999a,b)
NorESM	Modal	5 (nucleation and Aitken particles not included)	Storelvmo et al. (2008)
PMCAMx and GATOR	Sectional	10	Jacobson et al. (1996, 1997a,b)
Polair3D/Siream	Sectional	user specified	Jacobson et al. (1996, 1997a,b)
RegCM	Sectional	Coupling of MOSAIC in development	Zaveri et al. (2008)
WRF/Chem with MOSAIC	Sectional	4 or 8 bins	Fast et al. (2006), Shrivastava et al. (2011), Zaveri et al. (2008)
WRF-Chem-MADRID, CMAQ-MADRID	Sectional	8	Zhang et al. (2010a)

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Table 8. Radiative schemes (including short-wave, long-wave and photolysis modules which are considering gas [G], aerosol [A] and cloud water [C] effects) and ways of their coupling in selected online coupled models.

Model	Shortwave (SW)	Long-wave (LW)	Photolysis (PH)	Coupling step
BOLCHEM Mircea et al. (2008)	G: Climatology A: Calculation of local (grid-scal) optical properties for 5 dry aerosol types (SO_4, NH_4, Organic and Elemental Carbon, Dust) in three modes (Aitken, accumulation and coarse) and correction for aerosol water content. C: Cloud fraction and liquid/ice water content, at every level, from the prognostic cloud scheme.	G: Climatology A: Same as for SW C: Cloud fraction and liquid/ice water content, at every level, from the prognostic cloud scheme.	Photolysis rates are computed as a clear sky climatology modified locally by a grid-scale factor computed from the ratio between clear-sky and water content modified shortwave radiation. This factor accounts for the actual composition (gas, aerosol, water) seen by the model (see shortwave column on the left).t number	SW, LW, PH: User-defined; typically every 4 model time steps
COSMO-ART Vogel et al. (2009), Bangert et al. (2011)	G: None A: scattering and absorption by aerosols, depending on aerosol size distribution and chemical composition (all aerosol types), pre-computed lookup-tables (Mie calculations) C: Cloud optical properties based on effective radii of cloud droplets and ice crystals affected by aerosols acting as CCN, and by soot and dust acting as IN.	G: O_3 (climatology), CO_2 (clim), H_2O A: scattering and absorption by aerosols, depending on aerosol size distribution and chemical composition (all aerosol types), pre-computed lookup-tables (Mie calculations) C: Cloud optical properties based on effective radii of cloud droplets and ice crystals affected by aerosols acting as CCN, and by soot and dust acting as IN.	G: O_3 (climatology) A: Photolysis rates scaled proportional to SW radiation C: Photolysis rates scaled proportional to SW radiation.	SW, LW, PH: User-defined, typically every 15 min
COSMO-LM-MUSCAT Wolke et al. (2004a, 2012), Renner and Wolke (2010), Heinold et al. (2007, 2008, 2009), Helmert et al. (2007)	G: None A: Direct and semi-direct aerosol effect. As regards mineral dust: – Modified COSMO radiation scheme (Ritter and Geleyn, 1992), considering variations in the modelled size-resolved dust load. – Bin-wise offline Mie calculations of spectral optical properties using dust refractive indices from Sokolik and Toon (1999). For Biomass burning smoke ($PM_{2.5}$), as with the dust radiative feedback, but using mass extinction efficiency from Reid et al. (2005) for the computation of smoke optical thickness (Heinold et al., 2011a,b) Anthropogenic aerosol (EC, primary organic particles, NH_4NO_3, H_2SO_4, $(NH_4)_2SO_4$) is treated as dust but using mass extinction efficiencies from Kinne et al. (2006) to compute the optical thickness for each species. In addition, external mixing of the different components is assumed (Meier et al., 2012). C: None	G: None A: Direct and semi-direct aerosol effect. As regards mineral dust: – Modified COSMO radiation scheme (Ritter and Geleyn, 1992), considering variations in the modelled size-resolved dust load. – Bin-wise offline Mie calculations of spectral optical properties using dust refractive indices from Sokolik and Toon (1999). For Biomass burning smoke ($PM_{2.5}$), as with the dust radiative feedback, but using mass extinction efficiency from Reid et al. (2005) for the computation of smoke optical thickness (Heinold et al., 2011a,b) Anthropogenic aerosol (EC, primary organic particles, NH_4NO_3, H_2SO_4, $(NH_4)_2SO_4$) is treated as dust but using mass extinction efficiencies from Kinne et al. (2006) to compute the optical thickness for each species. In addition, external mixing of the different components is assumed (Meier et al., 2012). C: None	G: None A: None C: Modification of "clear sky" rates in dependence on the cloud cover or, alternatively, the liquid water pathway of the grid cells above.	SW, LW, PH: Coupling at every MUSCAT advection time step, given by time step control; COSMO radiation computation in separately-specified intervals of (usually) 1 h
ENVIRO-HIRLAM Baklanov et al. (2008), Korsholm et al. (2008)	G: Climatology – stratospheric O_3 A: Absorptance and transmittance calculation for 10 GADS aerosol types: insoluble, water soluble, soot, sea salt (acc. and coa. modes), mineral (nuc./acc./coa. modes), mineral (transported), sulfate droplets C: Grid and sub-grid scale bulk	G: H_2O, GHGs by constants A: Absorptance and transmittance calculation for 10 GADS aerosol types: insoluble, water soluble, soot, sea salt (acc. and coa. modes), mineral (nuc./acc./coa. modes), mineral (transported), sulfate droplets C: Grid and sub-grid scale bulk	G: None A: Only through temperature change caused by LW/SW C: Grid and sub-grid scale bulk	SW, LW, PH: Model time step

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Table 8. Continued.

Model	Shortwave (SW)	Long-wave (LW)	Photolysis (PH)	Coupling step
GEM Kaminski et al. (2008)	Correlated K (Li and Barker, 2005) with O_3, H_2O and aerosols from chemistry	Correlated K (Li and Barker, 2005) with O_3, H_2O and aerosols from chemistry	G,A,C all taken into account in J-value calculations using method of Landgraf and Crutzen (1998)	SW, LW, PH: model time step
IFS-MOZART (MACC/ECMWF) Flemming et al. (2009), Lindner et al. (2000), Fu (1996), Fu et al. (1998), Slingo 1989	G: Climatology based on the MACC-reanalysis A: Climatology or first direct effect and indirect effect, climatology is default. C: Various parameterisations	G: Climatology based on the MACC-reanalysis A: Climatology or first direct effect and indirect effect, climatology is default. C: Various parameterisations	G: Overhead O_3 A: None C: Simple shading parameterisation,	SW, LW, PH: 1 h
NMMB/BSC-CTM (BSC-CNS) Perez et al. (2011)	G: Climatology A: compute extinction efficiency for each mineral dust sectional bin and wavelength (Direct Aerosol Effect) C: Grid and sub-grid scale bulk	G:Climatology A:compute extinction efficiency for each mineral dust sectional bin and wavelength (Direct Aerosol Effect) C: Grid and sub-grid scale bulk	G: None A: None C: Bulk water content	SW, LW, PH: User-defined (typically every hour)
MCCM Grell et al. (2000), Forkel and Knoche (2006)	G: None A: Bulk total dry mass and aerosol water, fixed "typical" mass extinction coefficient for dry aerosol C: Coud droplet number: grid scale bulk only	G: None A: None C: Grid scale bulk	G: O_3 A: Bulk total dry mass and aerosol water, fixed "typical" mass extinction coeff. for dry aerosol C: Cloud droplet number: grid scale bulk only	SW, LW, PH: User-defined
MEMO/MARS-aero Moussiopoulos et al. (2012), Halmer et al. (2010), d'Almeida et al. (1991)	G: Constant background A: OPAC PM, PNC for water-soluble aerosols, averaged extinction coefficients for dry aerosol C: Cloud droplet number, parameterised profiles	G: Constant background A: OPAC PM, PNC for water-soluble aerosols, absorption + scattering coeff. for dry aerosol C: Cloud droplet number, parameterised profile	G: O_3 A: None C: None	SW, LW: User-defined PH: None
Meso-NH Lafore et al. (1998)	G: Climatology for O_3 (Fortuin and Langematz, 1994), constant background for CO_2, CH_4, N_2O, CFC11, CFC12 A: With prognostic aerosol (Tulet et al., 2005), radiative properties of aerosols according to Mie theory (Aouizerats et al., 2010). Without aerosol scheme, climatological aerosols. C: Effective radius calculated from the 2-moment microphysical scheme when explicitly used.	G: Climatology for O_3 (Fortuin and Langematz, 1994), constant background for CO_2, CH_4, N_2O, CFC11, CFC12 A: climatological aerosols. C: Effective radius calculated from the 2-moment microphysical scheme when explicitly used.	G: O_3 climatology A: aerosols climatology C: LWC when coupled on-line (1-D simulations) vs. Parametrisation of cloud impact (Chang et al., 1987) for 3-D simulations.	SW, LW, PH: User-defined
MetUM (MetOUnified Model) O'Connor et al. (2013), Savage et al. (2013)	G: N_2O, O_3, CH_4, depending on configuration – N_2O is not prognostic in AQ simulations A: Ammonium sulphate, ammonium nitrate, fossil-fuel BC & OC, mineral dust, biomass-burning, sea salt; based on mass & assumptions about hygroscopic growth and optical properties (see Bellouin et al., 2011) C: droplet number parameterised based on aerosol concentrations	G: N_2O, O_3, CH_4, depending on configuration – N_2O is not prognostic in AQ simulations A: Ammonium sulphate, ammonium nitrate, fossil-fuel BC & OC, mineral dust, biomass-burning, sea salt; based on mass and assumptions about hygroscopic growth and optical properties (see Bellouin et al., 2011) C: droplet number parameterised based on aerosol concentrations.	G: None (O_3 from climatology) A: Ammonium sulphate only C: Optical depth calculated based on cloud liquid water content only	SW, LW: User-defined (in AQ simulations typically every hour) PH: Every timestep

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Table 8. Continued.

Model	Shortwave (SW)	Long-wave (LW)	Photolysis (PH)	Coupling step
M-SYS (METRAS online version) von Salzen and Schlünzen (1999a)	G: Water vapour, O_3 A: Climatology prescribed C: Liquid water content	G: Water vapour, CO_2 A: Climatology prescribed C: Liquid water content	G: Standard atmosphere A: Climatology prescribed C: Direct dependence on liquid water content (in test, Uphoff, 2013)	SW, LW: Every minute PH: User defined, mostly every hour
RACMO2/LOTOS-EUROS, van Meijgaard et al. (2008), Schaap et al. (2008)	G: RRTM, H_2O + climatology CO_2, N_2O, SO_2, O_3, CFC11, CFC12 A: RRTM, Direct aerosol effect (dust, sea salt, black carbon, primary anthropogenic, sulfate, nitrate, ammonium), C: CCN based on sea salt, sulfate, nitrate concentrations (Menon 2004). Spectrally resolved subgrid scale, using cloud water path and cloud fraction	G: RRTM, H_2O + climatology CO_2, N_2O, SO_2, O_3, CFC11, CFC12 A: RRTM, no scattering, aerosol from climatology C: Spectrally resolved subgrid scale, using cloud water path and ice path, cloud fraction	G: CBM-IV (O_3, NO_2 etc.), A: None C: Cloud cover attenuation factor (bulk per grid cell) combined with Roeths flux (clear sky radiation)	Exchange of meteo and aerosol fields every 3 h, interpolation to hourly values SW: Every internal model time step, using hourly transmissivity update and solar angle per model timestep LW: Every internal time step, using hourly emissivity update and temperature profile per model time step PH: hourly update of photolysis rate based on meteorology and solar angle
RegCM4-Chem Zakey et al. (2006), Somon et al. (2006), Shalaby et al. (2012)	G: Gas climatology, except O_3 which can be interactive. A: Sulphate, fossil-fuel BC & OC, mineral dust, biomass-burning BC & OC, sea salt; based on mass & assumptions about hygroscopic growth, OPAC optical properties C: Coupling with prognostic microphysics in development	G: Gas climatology, except O_3 which can be interactive. A: Aerosol emission/absorption. C: None	G: None A: None C: Cloud OD effects on photolysis coefficients.	SW, LW: User-defined PH: Chemical time step, user-defined (typically 900 s).
RAMS/ICLAMS Kallos et al. (2009), Solomos et al. (2011)	G: Harrington radiation scheme with H_2O, O_3 and CO_2 and other gases; RRTMg OPAC A: Option to use Harrington radiation scheme with simulated natural aerosols and anthropogenic sulphates; RRTMg OPAC PM, option for online treatment of natural aerosols C: Interaction with explicitly solved liquid and ice hydrometeor size-spectra	G: Harrington radiation scheme with H_2O, O_3 and CO_2 and other gases; RRTMg OPAC A: Option to use Harrington radiation scheme with simulated natural aerosols and anthropogenic sulphates; RRTMg OPAC PM, option for online treatment of natural aerosols C: Interaction with explicitly solved liquid and ice hydrometeor size-spectra	G,A,C: Photolysis rates are computed online according to Madronich et al. (1987). Absorption cross sections and quantum yields according to Carter (2000).	SW, LW: User-defined PH: Embedded
WRF/Chem Grell et al. (2005, 2011), Fast et al. (2006), Zhang et al. (2010a)	G: Only climatologies A: **Dhudia**: Bulk total dry mass, EC, and aerosol water, fixed typical mass extinction coefficients for dry aerosol **GSFCSW**: Aerosol optical depth C: Cloud droplet number: considered RRTMG: uses aerosol optical properties from complex optical calculation module (MIE calculations)	G: Only climatologies A: **RRTM**: None C: RRTMG, CAM use aerosol optical properties and explicit cloud microphysics	G: O_3 A: **TUV**: like MCCM **Fast-J**: depending on simulated composition and size distribution (available for all aerosol modules) **F_TUV**: Acc. mode masses of EC, organic, NO_3, NH_4, SO_4, sea salt (for modal aerosol and bulk aerosols only) C: Cloud droplet number, bulk only	SW: User-defined LW: Equal to SW PH: User-defined
WRF-CMAQ Coupled System Pleim et al. (2008), Mathur et al. (2010), Wong et al. (2012)	RRTMG or CAM G: Constant background A: 5 groups: water-soluble, insoluble, sea-salt, BC and water; Direct aerosol effect only C: Scattering and absorption of cloud water droplet (parameterised)	RRTMG or CAM G: constant background A: 5 groups: water-soluble, insoluble, sea-salt, BC and water; Direct aerosol effect only C: Scattering and absorption of cloud water droplet (parameterised)	G: from the look-up table A:from the look-up table C: Uses a parameterization to correct the clear-sky photolysis rates for cloud cover	SW: User-defined LW: User-defined PH: N/A

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Table 9. Abbreviations and acronyms used in this article.

ABL	Atmospheric Boundary Layer
ACRANEB	Radiation scheme used in HARMONIE model (Ritter and Geleyn, 1992)
ADOM	Acid Deposition and Oxidant Model
AERO3	3rd generation CMAQ aerosol module
AERO5	5rd generation CMAQ aerosol module
AERO6	6rd generation CMAQ aerosol module
AIRS	The Atmospheric Infrared Sounder (instrument on board the NASA Aqua satellite)
ALADIN	Aire Limitée (pour l') Adaptation dynamique (par un) Développement InterNational (model and consortium)
AOD	Aerosol Optical Depth
AQ	Air Quality
AQMEII	Air Quality Model Evaluation International Initiative
AQUM	Air Quality in the Unified Model: limited area forecast configuration of the UK Met Office Unified Model which uses the UKCA (UK Chemistry and Aerosols) sub-model
AROME	Application of Research to Operations at Mesoscale-model (Météo-France)
ARW	The Advanced Research WRF solver (dynamical core)
BEIS3	Biogenic Emission Inventory System
BOLAM	Meteorological hydrostatic limited area model developed at CNR-ISAC in Bologna (IT)
BOLCHEM	Bologna limited area model for meteorology and chemistry (based on the BOLAM MetM)
BSC	Barcelona Supercomputing Center
BSC-CNS	Barcelona Supercomputing Center-Centro Nacional de Supercomputación
CAC	Chemistry-Aerosol-Cloud model (tropospheric box model)
CACM	Caltech Atmospheric Chemistry Mechanism
CAF	Coarray Fortran
CAM	The NCAR Community Atmospheric Model (CAM) Radiation Scheme
CAMx	Comprehensive Air quality Model with extensions
CAMx-AMWFG	Comprehensive Air Quality Model with Extensions – The Atmospheric Modeling and Weather Forecasting Group
CB-IV	Carbon Bond IV (chemistry module)
CBM-IV	The modified implementation of the Carbon Bond Mechanism version IV
CBM-Z	CBM-Z extends the CBM-IV to include reactive long-lived species and their intermediates, isoprene chemistry, optional DMS chemistry
CB05	The 2005 update to the gas-phase Carbon Bond mechanism (Yarwood et al., 2005)
CBR	The **C**uxart – **B**ougeault – **R**edelsperger turbulence closure scheme
CCM3	NCAR Community Climate Model (now Community Atmosphere Model – CAM)
CCN	Cloud Condensation Nuclei
CCTM-CMAQ	Chemistry-Transport Model of the CMAQ model
CDNC	Cloud Droplet Number Concentration
CDA	Chemical Data Assimilation
CHIMERE	A multi-scale CTM for air quality forecasting and simulation
C-IFS	Chemistry module for ECMWF IFS
CISL	Cell-integrated semi-Lagrangian (transport scheme)
CLASSIC	The Coupled Large-scale Aerosol Simulator for Studies In Climate (CLASSIC) aerosol scheme in MetUM
CMAQ	Community Multiscale Air Quality Modelling System (US Environmental Protection Agency)
CMAQ-MADRID	CMAQ-Model of Aerosol Dynamics, Reaction. Ionization, and Dissolution
CNR-ISAC	Institute of Atmospheric Sciences and Climate of the Italian National Research Council
COPS	Convective and Orographically-induced Precipitation Study
COST	European Cooperation in Science and Technology (http://www.cost.eu/)
COSMO	Consortium for Small-Scale Modelling (LAM model formerly called LM)
COSMO-ART	COSMO + Aerosols and Reactive Trace gases
COSMO-MUSCAT	COSMO + Multi-Scale Chemistry Aerosol Transport (model)
COT	Cloud Optical Thickness
CPU	Central Processing Unit
CTM	Chemistry-Transport Model
CWF	Chemical Weather Forecasting
CWFIS	Chemical Weather Forecasting and Information System
DMAT	Dispersion Model for Atmospheric Transport
DMI	Danish Meteorological Institute

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Table 9. Continued.

DREAM	Dust Regional Atmospheric Model
DWD	German Meteorological Service
ECMWF	European Centre of Medium-Range Weather Forecasts
ECHAM5/6-HAMMOZ	Global GCM ECHAM (version 5/6) + Aerosol chemistry and microphysics package HAM with additional parameterisations for aerosol–cloud interactions + the atmospheric chemistry model MOZART (MPI for Meteorology, Hamburg)
ECHAM5-HAM	Global aerosol–climate model
ECHAM/MESSy	Atmospheric Chemistry (EMAC) Numerical chemistry and climate simulation system
EEA/MDS	European Environment Agency/Model Documentation System
EM	Europa-Modell (Former DWD's hydrostatic meso-alpha scale regional NWP model
EMEP	European Monitoring and Evaluation Programme
EnKF	Ensemble Kalman filter
Enviro-HIRLAM	HIgh Resolution Limited Area Model HIRLAM with chemistry (DMI)
EQUISOLV	II Atmospheric gas-aerosol equilibrium solver
ESCOMPTE	Expérience sur Site pour COntraindre les Modèles de Pollution atmosphérique et de Transport d'Emissions (Urban boundary layer experiment
EuMetChem	The COST Action ES1004 – European framework for online integrated air quality and meteorology modelling
EURAD	European Air Pollution Dispersion model
ETA	The ETA MetM (uses the Eta vertical coordinate), originally developed in the former Yugoslavia (Mesinger et al., 1988), it is the old version of the WRF-model
ETEX	European Tracer Experiment
FARM	Flexible Air quality Regional Model
FCT	Flux-corrected transport advection scheme
GAMES	Gas Aerosol Modelling Evaluation System
GATOR	Gas, Aerosol, TranspOrt, Radiation AQ model (Stanford University)
GATOR-MMTD	GATOR – mesoscale meteorological and tracer dispersion model (also called GATORM)
GAW	Global Atmosphere Watch (WMO Programme)
GCM	General Circulation Models
GEM	Global Environmental Multiscale model (Canadian Meteorological Centre NWP)
GEM-AQ	GEM- + air quality processes online
GEMS	Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data
GEOS-Chem	GEOS–Chem is a global 3-D chemical transport model (CTM) for atmospheric composition driven by meteorological input from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office
GESIMA	German non-hydrostatic modelling community
GHG	Greenhouse gases
GLOMAP	GLobal Model of Aerosol Processes
GME	Global Model of DWD (DWD – German Weather Service)
GMES	Global Monitoring for Environment and Security
GOME	Nadir-scanning ultraviolet and visible spectrometer for global monitoring of atmospheric Ozone (on-board ERS-2)
GPU	Graphical Processing Units
GRAALS	radiation scheme to calculate vertical profiles of SW and LW radiative fluxes
GURME	GAW Urban Research Meteorology and Environment Project
HAM	Simplified global primary aerosol mechanism model
HARMONIE	Hirlam Aladin Research on Meso-scale Operational NWP in Europe (model)
HIRLAM	HIgh Resolution Limited Area Model (http://hirlam.org/)
HPC	High Performance Computing
IASI	Infrared Atmospheric Sounding Interferometer (onboard EUMETSAT METOP-A and then METOP-B satellite)
IC	Initial Conditions
ICLAMS	Integrated Community Limited Area Modeling System
IFS	Integrated Forecast System (ECMWF)
IN	Ice Nuclei
ISAC	Institute of Atmospheric Sciences and Climate (Italian National Research Council – CNR)
ISORROPIA	Thermodynamic aerosol model
JPL-06	Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies (NASA Jet Propulsion Laboratory Publication 06-2)
JRC-ENSEMBLE	The Joint Research Centre platform for model evaluation

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Table 9. Continued.

KIT	The Karlsruhe Institute of Technology
KPP	Kinetic Pre-Processors
LAI	Leaf Area Index
LAM	Limited Area Model
LAPS	Local Analysis and Prediction System
LES	Large Eddy Simulation
LMCSL	Locally Mass Conserving Semi-Lagrangian schemes (LMCSL-LL and LMCSL-3D)
LOTOS-EUROS	LOng Term Ozone Simulation – EURopean Operational Smog model
LW	Long-wave radiation
M7	Modal aerosol model
MACC	Monitoring Atmospheric Composition and Climate (EU project)
MADE	Modal Aerosol Dynamics model for Europe
MADE-SORGAM	Modal Aerosol Dynamics model for Europe (MADE) with the Secondary Organic Aerosol Model
MADEsoot	Modal aerosol module
MADRID	Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution
MARS	Model for the Atmospheric Dispersion of Reactive Species
MATCH	Multi-scale Atmospheric Transport and Chemistry Model
MCCM	Multiscale Climate Chemistry Model
MCM	Master Chemical Mechanism
MC2	Mesoscale Compressible Community (Canadian nonhydrostatic atmospheric model for Finescale Process Studies and Simulation)
MC2-AQ	MC2 with air quality modelling
MECCA	Revised MECCA1 (includes Aerosol chemistry submodule)
MECCA1	Module Efficiently Calculating the Chemistry of the Atmosphere (multi-purpose atmospheric chemistry model)
MECTM	MEsoscale Chemistry Transport Model
MEGAN	Model of Emissions of Gases and Aerosols from Nature (Guenther et al., 1993, 1995, 2006, 2012)
MEGAPOLI	Megacities: Emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation
MELCHIOR	Gas phase chemistry mechanism
MEMO	Eulerian non-hydrostatic prognostic mesoscale model (Aristotle University of Thessaloniki in collaboration with University of Karlsruhe)
MEMO/MARS	MEMO + photochemical dispersion model MARS
MARS-aero	Chemistry-transport model for reactive species including four chemical reaction mechanisms for the gaseous phase, with calculation of secondary aerosols, organic and inorganic
MESIM	Mesoscale Sea Ice Model
MESO-NH	Non-hydrostatic mesoscale atmospheric model (French research community)
MesoNH-C	Mesoscale Nonhydrostatic Chemistry model (coupled dynamics and chemistry)
MESOSCOP	Mesoscale flow and Cloud Model Oberpfaffenhofen (3d model for simulating mesoscale and microscale atmospheric processes)
MESSy	Modular Earth Submodel System
MetChem	Meteorology-Chemistry
MetM	Meteorological prediction model
METRAS	MEsoscale TRAnsport and fluid (Stream) model
MetUM	UK Met Office Unified Model
MICTM	MIcroscale Chemistry Transport Model
MITRAS	MIcroscale TRAnsport and fluid (Stream) model
MIPAS	Michaelson Interferometer for Passive Atmospheric Sounding (Fourier transform infra-red spectrometer on the ENVISAT-1 space mission)
MLS	Microwave Limb Sounder (on board NASA Earth Observing System Aura satellite)
MM5	Fifth Generation PSU/NCAR Mesoscale Model
MM5-CAMx	MM5 – Comprehensive Air quality Model with extensions
MM5-CHIMERE	MM5 – CHIMERE
MM5-CHEM	MM5 + chemistry module
MM5-CMAQ	Fifth Generation PSU/NCAR Mesoscale Model – Community Multiscale Air Quality Model
MOCAGE	Modèle de Chimie Atmosphérique à Grande Echelle
MOPITT	Measurements of Pollution in the Troposphere **(**on board NASA Terra satellite)
MOSAIC	Model for Simulating Aerosol Interactions and Chemistry
MOZAIC	**M**easurement of **Oz**one and water vapor by **A**irbus in-service air**C**raft
MOZART	Model for Ozone And Related Tracers (global CTM)

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Table 9. Continued.

MOZART2, 3 and 4	Model for Ozone And Related Tracers, version 2, 3, 4
MPI	Message Passing Interface
MRF	Markov random field (diffusion scheme)
M-SYS	Multiscale Model System consisting of components METRAS/MESIM, MITRAS, MECTM, MICTM
MUSCAT	Multi-Scale Chemistry Aerosol Transport model
NALROM	NOAA Aeronomy Lab Regional Oxidant Model
NAME	Numerical Atmospheric-dispersion Modelling Environment
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
NMMB	Nonhydrostatic Multiscale Meteorological Model on the B grid
NMMB/BSC-CTM	NMMB/BSC Chemical Transport Model
NMMB/BSC-Dust	online dust model within the global-regional NCEP/NMMB NWP-model
NRT	Near-Real Time
NWP	Numerical Weather Prediction
OCMC	Online Coupled Meteorology-Chemistry
OI	Optimal interpolation
OMI	Ozone Monitoring Instrument (on board Aura satellite)
OPAC	Optical Properties of Aerosols and Clouds (software library module)
OPANA	Operational version of Atmospheric mesoscale Numerical pollution model for urban and regional Areas
Open	MP Open Multi-Processing
ORILAM	Three-moments aerosol scheme
ORISAM	Sectional aerosol model
PAR	Photosynthetically Active Radiation
PD-FiTE	Partial Derivative Fitted Taylor Expansion (gas/liquid equilibria in atmospheric aerosol particles)
PEGASOS	EU FP7 project: Pan-European Gas-Aerosol-Climate interaction study (http://pegasos.iceht.forth.gr/)
PM	Particulate Matter
PMCAMx	3-D CTM simulating mass concentration and chemical composition of particulate matter (PM), based on the Comprehensive Air-quality Model with Extensions (CAMx)
PNC	Particle Number Concentration
Polair3D/MAM	Coupled 3-D chemistry transport model Polair3D to the multiphase model MAM
PROMOTE	PROtocol MOniToring for the GMES Service Element
RACM	Regional Atmospheric Chemistry Mechanism
RACM2	RACM Version2
RACM-MIM	RACM with the MIM (Mainzer Isopren Mechanismus) isoprene mechanism
RADM	Regional Acid Deposition Model
RADM2	the 2nd generation Regional Acid Deposition Model Mechanism
RADMK	Gas-phase chemistry module
RAMS	Regional Atmospheric Modeling Systems
RAQ	Regional Air quality
RCA-GUESS	A model of the coupled dynamics of climate, vegetation and terrestrial ecosystem biogeochemistry for regional applications (SMHI)
RCG REM3-CALGRID	Regional Eulerian Model – California Grid Model
RCM	Regional Climate Model
RegCM4	Regional Climate Model system (version4)
ReLACS	Regional Lumped Atmospheric Chemical Scheme
REMO	Regional Model
REMOTE	Regional Model with Tracer Extension
RK3	Runge-Kutta of 3rd order (Horizontal advection time splitting scheme)
RRSQRT	Reduced-rank square root Kalman filter
RRTM	Rapid radiative transfer model (retains the highest accuracy relative to line-by-line results for single column calculations).
RRTMG	RRTM for GCM Applications (provides improved efficiency with minimal loss of accuracy for GCM applications)
SAPRC90	The Statewide Air Pollution Research Center, Version 1999 for gas-phase reaction mechanism for the atmospheric photooxidation (Carter, 1990)
SAPRC99	SAPRC Version 1999
SAPRC07TB	New version of the SAPRC mechanism
SBUV	Solar backscattered ultraviolet (to monitor ozone density and distribution in the atmosphere aboard NOAA satellite)

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Table 9. Continued.

SCIAMACHY	SCanning Imaging Absorption SpectroMeter for Atmospheric ChartographY (satellite spectrometer designed to measure sunlight, transmitted, reflected and scattered by the earth's atmosphere or surface aboard ESA's ENVISAT).
SEMA	Sectional Multi-component Aerosol Model
SILAM	Air Quality and Emergency Modelling System (Finnish Meteorological Institute)
SLCF	Short-lived Climate Forcers
SLICE	"Semi-Lagrangian Inherently Conserving and Efficient" scheme for mass-conserving transport on the sphere
SOA	Secondary Organic Aerosol
SORGAM	Secondary organic aerosol formation model
STRACO	Soft TRAnsition and Condensation (Cloud scheme)
SW	Short Wave radiation
TANSO	Thermal And Near Infrared Sensor for Carbon Observation (on board the greenhouse gases observing satellite GOSAT)
THOR	An integrated air pollution forecast and scenario management system (National Environmental Research Institute (NERI), Denmark)
TKE	Turbulent Kinetic Energy
TM5	Transport Model (version5) (3-D atmospheric chemistry-transport ZOOM model)
TNO	the Netherlands Organisation for Applied Scientific Research
TTD	Turbulent Thermal Diffusion
TUV	Tropospheric Ultraviolet and Visible (radiation model)
TVD	Total Variation Diminishing (discretization scheme)
UKCA	UK Chemistry and Aerosols model
USSR	Union of Soviet Socialist Republics
VBS	Volatility Basis Set (approach)
VOTALP	Vertical Ozone Transports in the ALPs campaign
WAF	Weighted Average Flux scheme
WMO	World Meteorological Organization
WRF	The Weather Research and Forecasting model (NCAR)
WRF-Chem	The Weather Research and Forecast (WRF) model coupled with Chemistry
3/4DVar	3 or 4-dimensional variational assimilation

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Table 10. Chemical species.

BC	Black carbon
BVOC	Biogenic volatile organic compounds
CFC	Chlorofluorocarbon compounds (e.g. $CFCl_3$ and CF_2Cl_2)
CH_4	Methane
CO	Carbon monoxide
CO_2	Carbon dioxide
DMS	Dimethyl sulphide
EC	Elemental carbon
HCHO	Formaldehyde
HNO_3	Nitric acid
NH_3	Ammonia
NO	Nitric oxide
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides ($NO + NO_2$)
NO_3	Nitrate
N_2O	Nitrous oxide
N_2O_5	Dinitrogen pentoxide
OA	Organic aerosols and secondary (SOA)
OC	Organic carbon
O_3	Ozone
OH	Hydroxyl radical
$PM_{2.5}$	Particulate matter with diameter smaller than 2.5 µm
PM_{10}	Particulate matter with diameter smaller than 10 µm
POA	Primary organic aerosol
SOA	Secondary organic aerosol
SO_2	Sulphur dioxide
VOC	Volatile organic compounds

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Table B1. List of effects of meteorology on chemistry.

Meteorological parameter	Effect on . . .	Model variables
temperature	chemical reaction rates	T, reaction rate coefficients
	biogenic emissions	BVOC emission rates, isoprene, terpenes, DMS, pollen
	aerosol dynamics (coagulation, evaporation, condensation)	aerosol number size distributions scattering and absorption coefficients PM mass and composition
temperature and humidity	aerosol formation, gas/aerosol partitioning	gas phase SO_2, HNO_3, NH_3 particulate NO_3^-, SO_4^{2-}, NH_4^+ VOCs, SOA
	aerosol water take-up, aerosol solid/liquid phase transition	PM size distributions, extinction coefficient, aerosol water content
SW radiation	photolysis rates	JNO_2, JO1D, etc.
photosynthetic active radiation	biogenic emissions	SW radiation BVOC emissions, isoprene & terpene conc.
cloud liquid water and precipitation	wet scavenging of gases and particles	wet deposition (HSO_{3-}, SO_{4-}, NO_{3-}, NH_{4-}, Hg), precipitation (rain and total precip) cloud liq. water path
	wet phase chemistry, e.g. sulphate production	SO_2, H_2SO_4, SO_4^{2-} in ambient air and in cloud and rain water
	aerosol dynamics (activation, coagulation) aerosol cloud processing	aerosol mass and number size distributions
soil moisture	dust emissions, pollen emissions	surface soil moisture dust and pollen emission rates
	dry deposition (biosphere and soil)	deposition velocities, dry deposition rates (e.g. O_3, HNO_3, NH_3)
wind speed	transport of gases and aerosols on- vs. offline coupling interval, transport in mesoscale flows, bifurcation, circulations, etc.	U, V, (W)
	emissions of dust, sea salt and pollen	U, V dust, sea salt and pollen emission rates
atmospheric boundary layer parameters	turbulent and convective mixing of gases and aerosols in ABL, intrusion from free troposphere, dry deposition at surface	T, Q, TKE, surface fluxes (latent and sensible heat, SW and LW radiation) deposition velocities, dry deposition fluxe(O_3, HNO_3, NH_3)
lightning	NO emissions	NO, NO_2, lightning NO emissions
water vapour	OH radicals	Q, OH, HO_2, O_3

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Table B2. List of effects of chemistry on meteorology.

Chemical parameter	Effect on . . .	Model variables
aerosols (direct effect)	radiation (SW scattering/absorption, LW absorption)	AOD, aerosol extinction, single scattering albedo, SW radiation at ground (up- and downward), aerosol mass and number size distributions, aerosol composition: EC (fresh soot, coated), OC, SO_4^{2-}, NO_3^-, NH_4^+, Na, Cl, H_2O dust, metals, base cations
aerosols (direct effect)	visibility, haze	aerosol absorption & scattering coefficients, RH, aerosol water content
aerosols (indirect effect)	cloud droplet or crystal number and hence cloud optical depth	interstitial/activated fraction, CCN number, IN number, cloud droplet size/number, cloud liquid and ice water content
aerosols (indirect effect)	cloud lifetime	cloud cover
aerosols (indirect effect)	precipitation (initiation, intensity)	precipitation (grid scale and convective)
aerosols (semi-direct effect)	ABL meteorology	AOD, ABL height, surface fluxes (sensible and latent heat, radiation)
O_3	UV radiation	O_3, SW radiation < 320 nm
O_3	thermal IR radiation, temperature	O_3, LW radiation
NO_2, CO, VOCs	precursors of O_3, hence indirect contributions to O_3 radiative effects	NO_2, CO, total OH reactivity of VOCs
SO_2, HNO_3, NH_3, VOCS	precursors of secondary inorganic and organic aerosols, hence indirect contributors to aerosol direct and indirect effects	SO_2, HNO_3, NH_3, VOC components (e.g. terpenes, aromatics, isoprene)
soot deposition on ice	surface albedo change	snow albedo

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Table B3. Observation data sets available for model evaluation (name, number of sites, frequency of measurements, preferred model output type).

Parameter	Evaluation datasets	# sites	Database	Frequency H – hourly D – daily W – weekly M – monthly I – irregular	Model output type LP – local profiles 2Dc – 2D column
$PM_{2.5}$	various techniques gravimetric, TEOM, etc.	835 50	EEA Airbase EMEP	D H, D, W	in-situ in-situ
PM_{10}	various techniques gravimetric, TEOM, etc.	3000 80	EEA Airbase EMEP	D H, D, W	in-situ in-situ
aerosol optical depth (AOD) and Angstrom exponent (ratio of AOD at different wavelengths)	AERONET AOD @ 443, 490, 555, 667 nm, Angström parameter MODIS (AOD, Angstr. exponent) CALIPSO	60–80 satellite satellite	AERONET MODIS CALIPSO	H twice D I	in-situ 2Dc LP
aerosol extinction, absorption and scattering coefficients	nephelometer, aethalometer AERONET single scattering albedo	10–15 60–80	EMEP, EUSAAR AERONET	H H	in-situ in-situ
aerosol size distribution	SMPS/DMPS	24	EMEP, EUSAAR	H	in-situ
aerosol composition (non-refractory PM_1)	aerosol mass spectrometry (AMS)	9 (campaigns)	EMEP	H	in-situ
aerosol elemental and organic carbon	EC/OC monitors, thermo-optical	18	EMEP, EUSAAR	D, W	in-situ
inorganic aerosol comp. NO_3^-, SO_4^{2-}, NH_4^+	filterpack, mini-denuders MARGA	90 2	EMEP MARGA	D H	in-situ in-situ
O_3	ozone monitor ozone monitor MOZAIC	3000 100 aircraft	EEA Airbase EMEP/EBAS MOZAIC	H H ~ D	in-situ in-situ LP

Network/database acronyms and websites:
AERONET, Aerosol Robotic Network, http://aeronet.gsfc.nasa.gov/
BSRN, Baseline Surface Radiation Network, http://www.bsrn.awi.de/
CALIPSO, Cloud-Aerosol Lidar and Infrared Pathfinder, http://www-calipso.larc.nasa.gov/
Cloudnet, http://www.cloud-net.org/index.html
EEA Airbase, Air quality database of European Environmental Agency, http://acm.eionet.europa.eu/databases/airbase/
EMEP, European Monitoring and Evaluation Program, http://www.emep.int/index.html, http://ebas.nilu.no
ESA CCI soil moisture, http://www.esa-soilmoisture-cci.org/
EUCAARI, European Integrated Project on Aerosol Cloud Climate Air Quality Interactions, http://www.atm.helsinki.fi/eucaari/
EUSAAR, European Supersites for Atmospheric Aerosol Research, http://www.eusaar.net/, http://ebas.nilu.no
GPCP, Global Precipitation Climatology Project, http://www.gewex.org/gpcp.html, http://gpcc.dwd.de
MARGA, Monitor for aerosols and gases in air, http://products.metrohm.com/prod-MARGA.aspx
MODIS, Moderate Resolution Imaging Spectroradiometer, http://modis.gsfc.nasa.gov/
RAOB, WMO radiosonde observations network, http://www.esrl.noaa.gov/raobs/
SYNOP, WMO surface meteorology network, http://www.wmo.int/pages/prog/www/ois/rbsn-rbcn/rbsn-rbcn-home.htm

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Table B3. Continued.

Parameter	Evaluation datasets	# sites	Database	Frequency H – hourly D – daily W – weekly M – monthly I – irregular	Model output type LP – local profiles 2Dc – 2D column
NO_2	NO_x monitors (significant interference from HNO_3, PAN)	3200	EEA Airbase	H	in-situ
	chemiluminescence, filterpack, abs. solution, etc.	85	EMEP	H, D	in-situ
	satellite NO_2 columns (OMI, GOME-2, SCIA)	satellite	TEMIS	~ D	2-D
CO	CO monitor	1300	EEA Airbase	H	in-situ
SO_2	SO_2 monitor	2000	EEA Airbase	H, D	in-situ
		90	EMEP	H, D	in-situ
HNO_3, NH_3	filterpack	90	EMEP	D, W	in-situ
	MARGA	2	MARGA	H	in-situ
OH radicals	selected ROx meas. (PERCA, LIF, open path)	–	–	–	in-situ
wet deposition: HSO_{3-}, SO_{4-}, NO_{3-}, NH_{4-}, Hg	EMEP wet deposition	90	EMEP	W	in-situ
dry deposition: O_3, HNO_3, NH_3, etc.	no routine observations, selected eddy flux campaigns	–	–	–	in-situ
precipitation	EMEP precipitation	90	EMEP	W	in-stiu
VOCs incl. isoprene	GC-MS, GC-FID	11	EMEP	twice W	in-situ
soot deposition on ice	MODIS (black sky) albedo	satellite	MODIS	8-daily	surface albedo
temperature, humidity, wind, pressure	SYNOP	1300	SYNOP	H	in-situ
	RAOB	100	RAOB	twice D	LP

Network/database acronyms and websites:
AERONET, Aerosol Robotic Network, http://aeronet.gsfc.nasa.gov/
BSRN, Baseline Surface Radiation Network, http://www.bsrn.awi.de/
CALIPSO, Cloud-Aerosol Lidar and Infrared Pathfinder, http://www-calipso.larc.nasa.gov/
Cloudnet, http://www.cloud-net.org/index.html
EEA Airbase, Air quality database of European Environmental Agency, http://acm.eionet.europa.eu/databases/airbase/
EMEP, European Monitoring and Evaluation Program, http://www.emep.int/index.html, http://ebas.nilu.no
ESA CCI soil moisture, http://www.esa-soilmoisture-cci.org/
EUCAARI, European Integrated Project on Aerosol Cloud Climate Air Quality Interactions, http://www.atm.helsinki.fi/eucaari/
EUSAAR, European Supersites for Atmospheric Aerosol Research, http://www.eusaar.net/, http://ebas.nilu.no
GPCP, Global Precipitation Climatology Project, http://www.gewex.org/gpcp.html, http://gpcc.dwd.de
MARGA, Monitor for aerosols and gases in air, http://products.metrohm.com/prod-MARGA.aspx
MODIS, Moderate Resolution Imaging Spectroradiometer, http://modis.gsfc.nasa.gov/
RAOB, WMO radiosonde observations network, http://www.esrl.noaa.gov/raobs/
SYNOP, WMO surface meteorology network, http://www.wmo.int/pages/prog/www/ois/rbsn-rbcn/rbsn-rbcn-home.htm

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Table B3. Continued.

Parameter	Evaluation datasets	# sites	Database	Frequency H – hourly D – daily W – weekly M – monthly I – irregular	Model output type LP – local profiles 2Dc – 2D column
SW and LW radiation at ground	global radiation (direct+diffuse), long-wave downward radiation	13	BSRN	H	in-situ
photolysis rates (J_{NO_2}, J_{O1D}, etc.)	no routine obs. available	–	–	–	–
precipitable water (water vapour column)	AERONET RAOB	60–80 100	AERONET RAOB	H twice D	in-situ, LP LP
boundary layer turbulence, TKE PBL height	selected tall tower and FLUXNET sites Radiosondes Select. Lidars/Ceilometers	85 100 –	FLUXNET RAOB –	H twice D –	in-situ LP, 2-D –
cloud cover, cloud top, cloud optical depth, cloud base	SEVIRI MODIS AVHRR CALIPSO Lidar Cloudnet Selected Lidars, Ceilometers	satellite satellite satellite satellite 3 –	 Cloudnet –	H twice D D I H H	2-D 2-D 2-D in-situ, LP in-situ
cloud liquid water path	AERONET Cloudnet	60–80 3	AERONET Cloudnet	H H	in-situ, LP in-situ, LP
soil moisture	satellite soil moisture	satellite	ESA CCI	D	

Network/database acronyms and websites:
AERONET, Aerosol Robotic Network, http://aeronet.gsfc.nasa.gov/
BSRN, Baseline Surface Radiation Network, http://www.bsrn.awi.de/
CALIPSO, Cloud-Aerosol Lidar and Infrared Pathfinder, http://www-calipso.larc.nasa.gov/
Cloudnet, http://www.cloud-net.org/index.html
EEA Airbase, Air quality database of European Environmental Agency, http://acm.eionet.europa.eu/databases/airbase/
EMEP, European Monitoring and Evaluation Program, http://www.emep.int/index.html, http://ebas.nilu.no
ESA CCI soil moisture, http://www.esa-soilmoisture-cci.org/
EUCAARI, European Integrated Project on Aerosol Cloud Climate Air Quality Interactions, http://www.atm.helsinki.fi/eucaari/
EUSAAR, European Supersites for Atmospheric Aerosol Research, http://www.eusaar.net/, http://ebas.nilu.no
GPCP, Global Precipitation Climatology Project, http://www.gewex.org/gpcp.html, http://gpcc.dwd.de
MARGA, Monitor for aerosols and gases in air, http://products.metrohm.com/prod-MARGA.aspx
MODIS, Moderate Resolution Imaging Spectroradiometer, http://modis.gsfc.nasa.gov/
RAOB, WMO radiosonde observations network, http://www.esrl.noaa.gov/raobs/
SYNOP, WMO surface meteorology network, http://www.wmo.int/pages/prog/www/ois/rbsn-rbcn/rbsn-rbcn-home.htm

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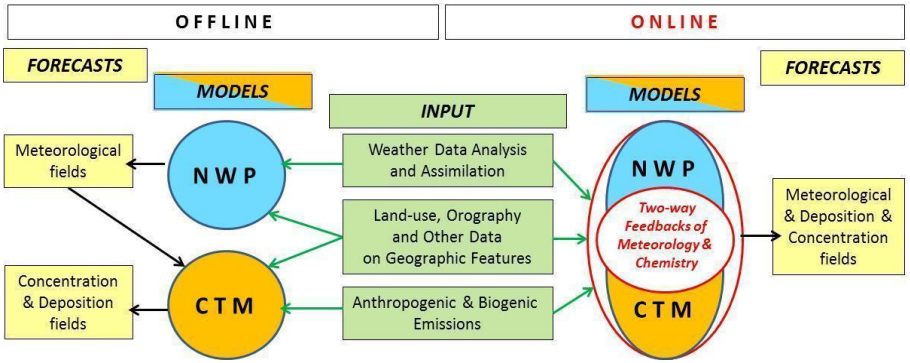

Fig. 1. Schematic diagram of (left) offline and (right) online coupled NWP and CTM modelling approaches for AQ and meteorology simulation.

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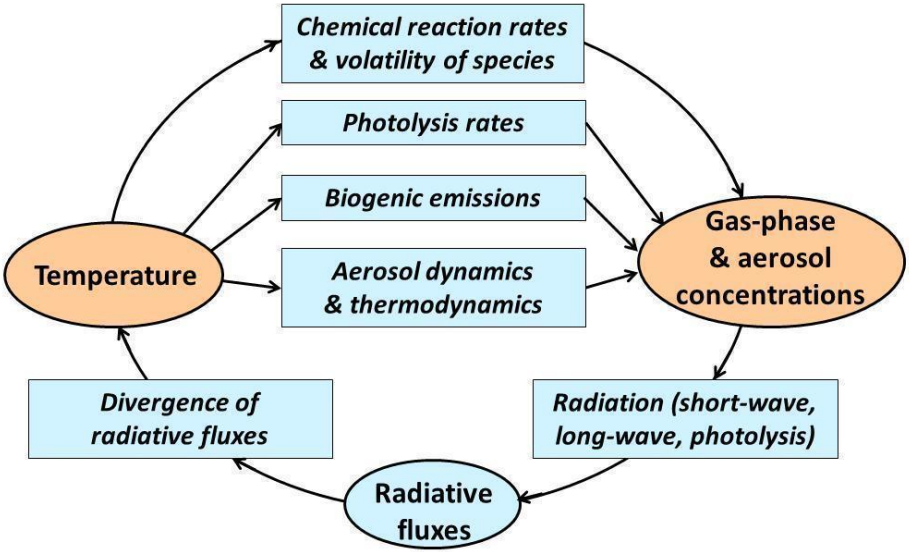

Fig. 2. Conceptual model of impacts from temperature on concentrations and vice versa.

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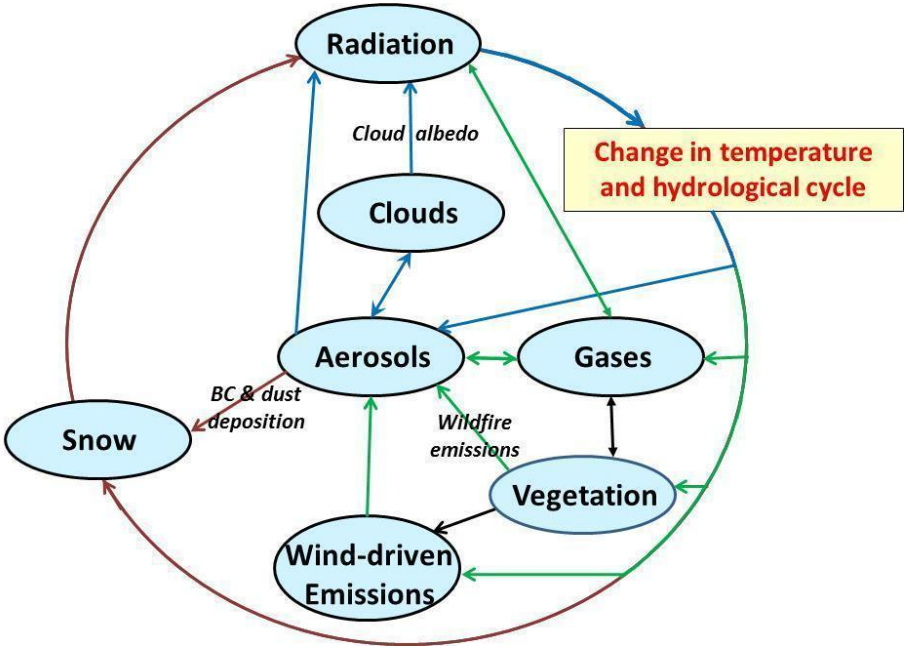

Fig. 3. Interactions between aerosols, gases and components of the Earth system.

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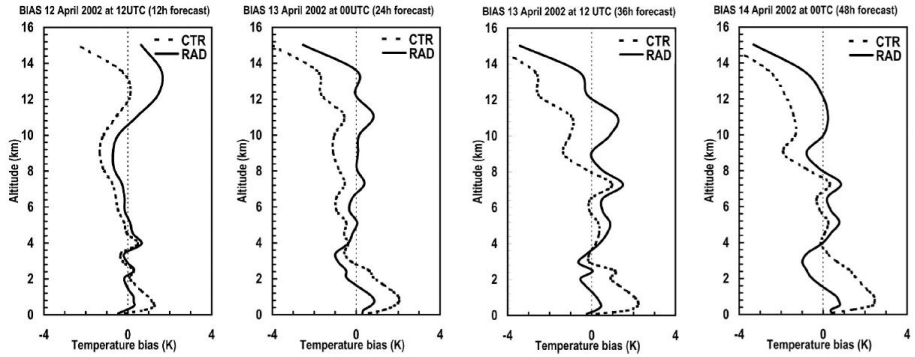

Fig. 4. Vertical profiles of the atmospheric temperature bias between a control run (CTR) without and a full run (RAD) with SW and LW radiative interaction of dust aerosols. Profiles are over an area most strongly affected by Saharan dust (30° N–45° N, 0° E–20° E) for the 12, 24, 38, and 48 h forecasts of the 00:00 UTC forecast cycle on 12 April 2002 (adopted from Pérez et al., 2006).

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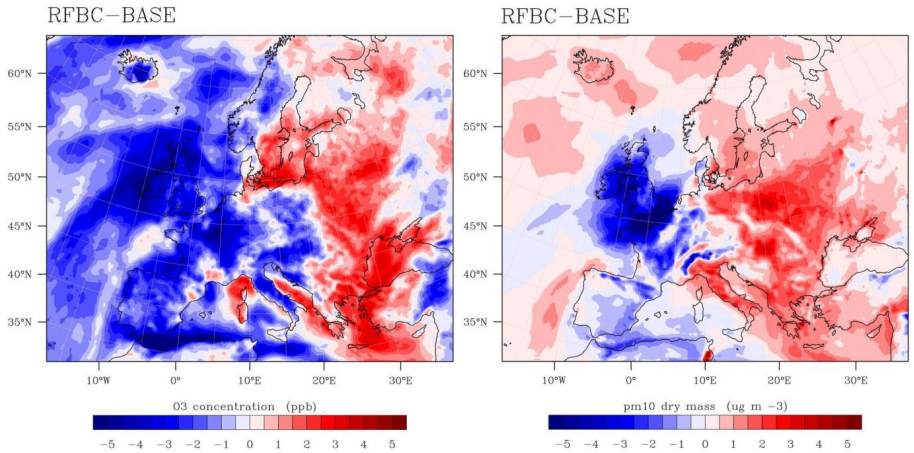

Fig. 5. Differences in ozone (left) and PM_{10} (right) concentrations in July 2006 between two WRF-Chem simulations. The BASE simulation does not consider interactions between aerosols and meteorology, whereas the RFBC simulation considers both direct and indirect effects (adopted from Forkel et al., 2012).

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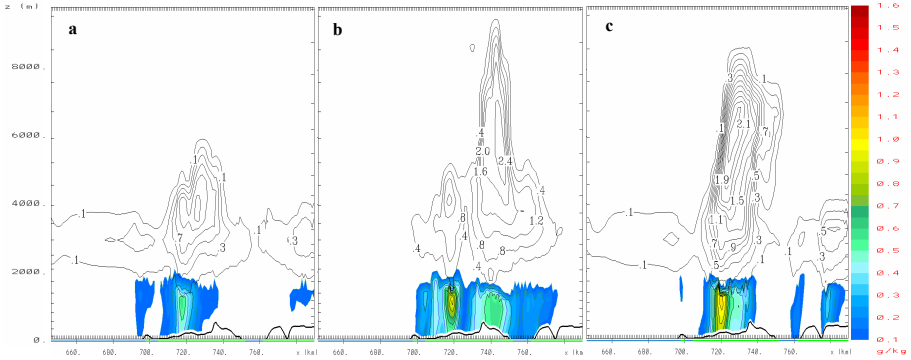

Fig. 6. West to East cross-section of rain mixing ratio (color palette in g kg^{-1}) and ice mixing ratio (black line contours in g kg^{-1}) at the time of highest cloud top over Haifa. **(a)** 09:00 UTC 29 January 2003 assuming 5 % hygroscopic dust. **(b)** 10:00 UTC 29 January 2003 assuming 20 % hygroscopic dust. **(c)** 09:00 UTC 29 January 2003 assuming 5 % hygroscopic dust and number of ice nuclei increased by a factor 10. Adopted from Solomos et al. (2011).

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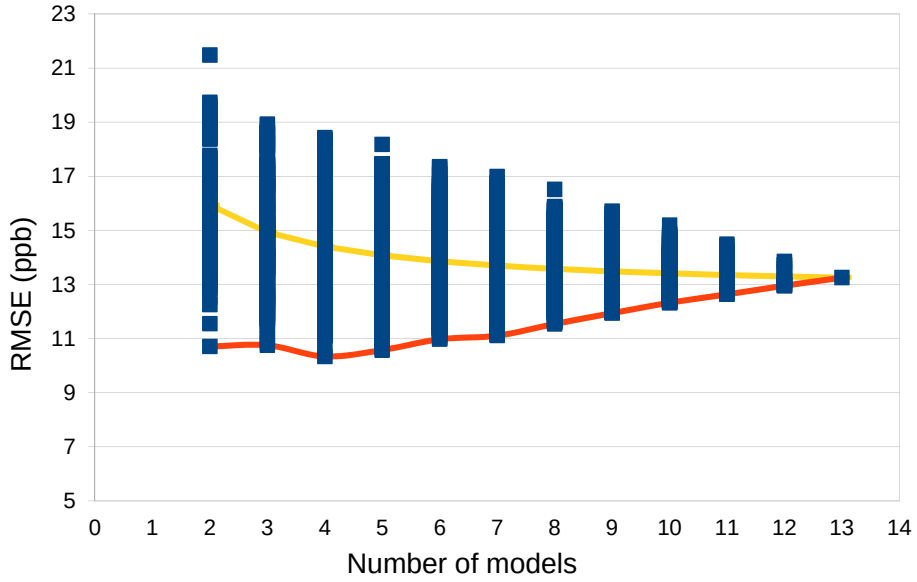

Fig. 7. Variability of root mean square error (RMSE) for ozone calculated from an ensemble of 13 regional scale models applied to a sub- region of Europe. The blue bars indicate the range of RMSE obtained for all combinations of 13 models combined in all possible pairs, triplets, quadruplets etc. The yellow curve represents the average RMSE produced by all possible combinations of 13 model result when grouped as groups of 2, 3, 4 up to 13 members. The red curve connects the minimum RMSE produced by the various combinations, thus indicating that an increasing number of results does not necessarily reduces the error and that there is an optimal combination of models which produced a minimum RMSE (from Solazzo et al., 2012a).

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