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A review of operational, regional-scale, chemical weather forecasting models in Europe

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Abstract. Numerical models that combine weather forecasting and atmospheric chemistry are here referred to as chemical weather forecasting models. Eighteen operational chemical weather forecasting models on regional and continental scales in Europe are described and compared in this article. Topics discussed in this article include how weather forecasting and atmospheric chemistry models are integrated into chemical weather forecasting systems, how physical processes are incorporated into the models through parameterization schemes, how the model architecture affects the predicted variables, and how air chemistry and aerosol processes are formulated. In addition, we discuss sensitivity analysis and evaluation of the models, user operational requirements, such as model availability and documentation, and output availability and dissemination. In this manner, this article allows for the evaluation of the relative strengths and weaknesses of the various modelling systems and modelling approaches. Finally, this article highlights the most prominent gaps of knowledge for chemical weather forecasting models and suggests potential priorities for future research

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directions, for the following selected focus areas: emission inventories, the integration of numerical weather prediction and atmospheric chemical transport models, boundary conditions and nesting of models, data assimilation of the various chemical species, improved understanding and parameterization of physical processes, better evaluation of models against data and the construction of model ensembles.

1 Introduction

Chemical weather is defined here as the short-term (less than two weeks) variability of the atmospheric chemical composition. This definition is complementary to the traditional meteorological definition of weather, which is commonly characterized only by physical variables (e.g. temperature, wind, mass, radiation, humidity). Methods that include a combination of weather forecasting and atmospheric chemistry simulations are here referred to as chemical weather forecasting (CWF). CWF can therefore be seen as a specific category of air-quality forecasting, where air-quality forecasting models using numerical weather prediction (NWP) models are CWF models, but air-quality forecasting models using statistical methods are not (Kukkonen et al., 2009c). Similarly, for accuracy and consistency in replacing the traditional term *air-quality forecasting and information system*, we introduce a new term *chemical weather forecasting and information system* (CWFIS) to represent the integrated system responsible for the prediction and dissemination of chemical weather forecasts.

In the literature, numerous other terms are also used to refer to chemical weather and air-quality forecasting systems, such as, for instance, "chemical weather and air-quality forecasting models", "regional, continental or global air-quality models", "dispersion models", "atmospheric chemistry models", "chemical transport models", "air-chemistry models" and "atmospheric chemistry transport models". In this article, we also use some of these terms; however, "chemical transport models" is used specifically to refer to the atmospheric chemistry simulations.

Sometimes the term biological weather forecasting is used to refer to forecasting of biological constituents in the air, such as various pollen species and airborne allergens. This paper does not specifically address biological weather forecasting, although some of the considered models include treatments for airborne pollen species.

All the acronyms used in this article have been listed at the end of this article.

Lawrence et al. (2005) have previously reviewed the then-current state of CWF and emerging research challenges. Baklanov et al. (2008a, 2010b) and Schluenzen and Sokhi (2008) summarized existing mesoscale modelling systems and capabilities as an initial step to formulate recommendations for a unified integrated framework for modelling systems, although they did not compare the mathematical architecture of the various modelling systems. Baklanov (2010) also presented some gaps in our current understanding and recommended directions of future research for integrated CWF systems, although a valuable addition would be a more comprehensive set of recommendations summarizing the most urgent gaps of knowledge and research needs.

There are currently tens, possibly more than a hundred, CWFISs on a local, regional and continental scale in Europe and worldwide. Although abundant literature exists on the properties of individual models, scientific articles presenting compilations or synthesis of this information are scarce. Furthermore, the scientific evaluation of models against data – defined to include also the detailed analysis and evaluation of the mathematical structure of such models or modelling systems in terms of the underlying physics and chemistry – are more limited. No scientific evaluations have been presented of a larger number of CWF models, although the Air-Quality Model Evaluation International Initiative (AQMEII) model intercomparison study is expected to be able to provide such information (Rao et al., 2011).

Despite a plethora of modelling options, it is far from obvious, which are the optimal ones in most cases. Thus, a systematic review of these options could substantially assist in evaluating the strengths and weaknesses of the various methods, and thus contribute to the development of better and more robust modelling methods in the future. Consequently, this present article aims to bring the field up to date with a comprehensive summary and assessment of the state of CWF in Europe.

1.1 European-wide projects on chemical weather modelling and forecasting

This study is part of the European Cooperation in Science and Technology (COST) ES0602 action, which provides a forum for benchmarking approaches and practices in data exchange and multi-model capabilities for CWF and near real-time information services in Europe (http://www.chemicalweather.eu). The action was initiated by the Network of European Meteorological services (EUMETNET, http://www.eumetnet.eu) and the European Environment Agency (EEA). The content of this COST action, its main objectives and organisation have been reviewed by Kukkonen et al. (2009a, b), and the main results by Kukkonen et al. (2009c). The COST action includes participants from 20 countries, and its duration is from 2007 to 2011.

The COST ES0602 action has constructed a European open-access CWF portal (ECWFP) that includes access to a substantial number (more than 20) of available chemical weather forecasting systems (CWFS) and their numerical forecasts; these cover in total 31 areas in Europe (Balk et al., 2011; http://www.chemicalweather.eu/Domains). This portal can be used to find out, which CWF services are available for a specific domain, for specific source categories or

for specific pollutants. Such a single point of reference for European CWF information has not previously been available. The Action has also investigated and reviewed existing chemical weather information systems and services (e.g. Karatzas and Kukkonen, 2009). This study has also been part of the EU-funded projects MEGAPOLI (Megacities: emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation, http://www.megapoli.info; Baklanov et al., 2010a) and TRANSPHORM (Transport-related Air Pollution and Health impacts – Integrated Methodologies for Assessing Particulate Matter, http://www.transphorm.eu/).

There are several other prominent ongoing European projects that address CWF. Some of the most important operational CWF programs lie within the EU-ESA (European Space Agency) programme GMES (Global Monitoring for Environment and Security, http://www.gmes.info), viz. the GEMS (Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data, http://gems.ecmwf.int) and PROMOTE (PROtocol MOniToring for the GMES Service Element, http://www.gse-promote.org; Poupkou et al., 2006) projects. The GMES Atmospheric Services focus on pre-operational monitoring and forecasting of atmospheric composition, dynamics and thermodynamics through advanced exploitation of satellite and in-situ data, on a European, national and local level. We define an operational modelling system as an automated one, which has a fall-back procedure in case the forecast fails. The latter is commonly a human part of the operational cycle. A pre-operational modelling system could be defined as an automated system with a fall-back procedure that is not comprehensive.

There are also other related EU-funded projects, such as CITYZEN (megaCITY – Zoom for the Environment, https://wiki.met.no/cityzen/start), EUCAARI (The European Integrated project on Aerosol Cloud Climate and Air-Quality Interactions, http://www.atm.helsinki.fi/eucaari) and EUSAAR (European Supersites for Atmospheric Aerosol Research, http://www.eusaar.net). Within the GEMS project, analyses and 72-h forecasts have been presented using 12 state-of-the-art regional chemical weather models on a pre-operational daily basis (http://gems.ecmwf.int). The models rely on the operational meteorological forecasts of the European Centre for Medium-Range Weather Forecasts (ECMWF), as well as on GEMS global chemical weather data. They all consider the same high-resolution (5-km horizontal grid spacing) anthropogenic and biogenic emissions inventories.

An example of a small-scale network of a few operational chemical weather services has been constructed within the first and second stages of the PROMOTE project. Although GEMS and PROMOTE CWF services have constituted major advances in this field – and are evidently valuable for a range of stakeholders – a limitation is that these projects have had a closed membership and have been fairly oriented around development. There is a need to involve additional stake-holders in a more comprehensive way, such as the national environmental agencies. This is also the task of the continuation projects of GEMS and PROMOTE, the MACC (Monitoring Atmospheric Composition and Climate) and PASODOBLE (Promote Air-Quality Services integrating Observations – Development Of Basic Localised Information for Europe) projects.

Another relevant program is Global Earth Observation and Monitoring (GEOmon, www.geomon.eu), the goal of which is to build an integrated European ground-based observational network of atmospheric composition to complement satellite observations. It lays the foundation for a European contribution to GEOSS (Global Earth Observation System of Systems, http://www.epa.gov/geoss) and optimizes the European strategy of monitoring atmospheric composition measurements (Tørseth and Fahre Vik, 2009).

1.2 Aims and scope of this study

Given the large variety of existing modelling systems and options, we must limit the scope of this article. Specifically, we select 18 operational CWF models on regional and continental scales (distance scales of approximately 10–6000 km) in Europe for more detailed analysis. These models are among the most widely used in Western European countries, as well as in Eastern and Central-Eastern European countries. However, this collection of models is by no means exhaustive. Moreover, some of these models have been mainly developed elsewhere, especially in the United States.

This paper has three main aims. The first aim is to gather information on the selected operational CWF models in a systematic and harmonized format. The second aim is to provide and synthesize information that makes it possible for the readers to evaluate the relative strengths and limitations of the various models, and the components of the modelling systems. However, it is not the goal of this study to rank the models, or advocate one model over another. The third aim is to highlight the most prominent gaps of knowledge in CWF and to suggest priorities for future research directions.

We do not address purely diagnostic models, which do not include forecasting capabilities. The emergency preparedness models (such as those developed in case of nuclear and chemical accidents) are also outside the scope of this study. Because this article focuses on regional-scale models, we do not address modelling on global or urban scales. This study also addresses only operational CWF models. In comparison with genuine research models, and versions of operational models that are used only for research purposes, such operational models can include simplifications, such as a reduced resolution, smaller domains, and less sophisticated physics and chemistry modules. However, the discussion of the detailed operational characteristics of the modelling systems is outside the scope of this study; these have been addressed by Balk et al. (2011). This article also does not contain any

new numerical intercomparisons of model predictions, or any novel evaluation of model predictions with data.

1.3 Organization of this article

This article is organized in the following manner. Section 2 introduces the 18 different CWF modelling systems and gives an overview of some of the relevant physical processes. Section 3 discusses the numerical weather prediction components of the models. Section 4 discusses the processes in the atmospheric dispersion and chemistry modelling components. Section 5 discusses the evaluation of the models. Section 6 discusses user operations of the model, including availability, computer requirements, documentation, user interfaces, sensitivity analyses, and dissemination. Section 7 looks forward to discuss emerging issues in the CWF community, including the identification of major gaps of knowledge and future research needs. Finally, Sect. 8 concludes this article.

The main characteristics of the selected 18 CWF models considered in this study have been summarised and intercompared in several tables. Overviews of the main properties of the CWF and NWP models are first presented in Tables 1 and 2. The atmospheric dispersion, chemistry and aerosol modelling, and deposition components are reviewed in Tables 3–7. The natural emissions, and the grid spacings and coordinate systems are presented in Tables 8 and 9. The evaluation of each CWF model, and the availability, user communities, documentation, and presentation of forecasts in the internet are presented in Tables 10–12. Finally, in relation to future research needs, adjoint (inverse) dispersion modelling is reviewed in Table 13.

2 Introduction to operational chemical weather forecasting models

This section addresses some key concepts and introduces the main physical and chemical processes that are relevant for CWF. We have selected 18 operational, regional and continental scale CWF modelling systems for a more detailed examination.

First, we explain how the models were selected and how the key information was derived (Sect. 2.1). Second, we address how numerical weather prediction models can be integrated with CWF models (Sect. 2.2). For readability, this section also contains an introduction on the selected main properties of those models (Sect. 2.3), before we present a more detailed analysis and inter-comparison of model treatments for specific processes.

2.1 Criteria for the selection of the models and the use of information sources

A fairly large number of models were first suggested by the participants of the COST ES0602 action for a more detailed examination; participants from more than 20 European countries (listed in Kukkonen et al., 2009c) were encouraged to volunteer for this activity. Finally, 18 models were selected for inclusion in this article.

The main criteria for the selection of the 18 models were (i) the prominence and wide usage of the models, and (ii) the sufficient availability of scientific literature and Web-based documentation on the relevant model properties. Most of the models addressed in this study are also contained in the Model Documentation System (MDS) of EEA, accessible at http://air-climate.eionet.europa.eu/databases/MDS/index_html, and in the joint COST 728 and COST 732 Model Inventory (C728/732MI), accessible at http://www.cost728.org (Schluenzen and Sokhi, 2008). We also aimed to present a balanced geographical representation across Europe.

Clearly, there are some prominent CWF systems that are not included in this article, such as CALIOPE (http://www.gmes-masters.com/service-application/caliope-system) and GEM-AQ (http://ecoforecast.eu/). Baldasano et al. (2008) and Pay et al. (2010) describe the Spanish CWF system CALIOPE and discuss modelling results over Europe and Spain. The CALIOPE model is operational and provides air-quality forecasts over Europe on a horizontal resolution of 12×12 km and over Spain on 4×4 km (http://www.bsc.es/caliope). The CALIOPE system also accounts for the mineral dust transport from North Africa to Europe and Spain by means of the BSC-DREAM8b model (Nickovic et al., 2001 and Perez et al., 2006), and gas-phase and aerosol pollutants by means of the CMAQ model.

To obtain the most credible and up-to-date information, model properties were derived primarily from published literature and from the developers or users of each model. We also used secondary information sources from the Web, such as the MDS, C728/732MI and the various web pages of individual modelling systems. In some cases, we received conflicting information from these sources. When that has happened, we extracted the information from the published literature whenever available, and then contacted the model developers for confirmation.

2.2 The integration and coupling of numerical weather prediction and chemical transport models

How NWP models couple with CWF models can be realized in one of two principal ways. Grell et al. (2005) and Baklanov et al. (2008a) suggested the following definitions.

Off-line modelling systems (also called one-way interactive models) contain a separate chemical transport model (CTM) driven by meteorological input data from meteorological pre-processors, measurements or diagnostic models, is driven by analysed or forecasted meteorological data from NWP archives or datasets, or reads output files from operational NWP models or specific meteorological models at limited time intervals (e.g. 1, 3, 6 h).

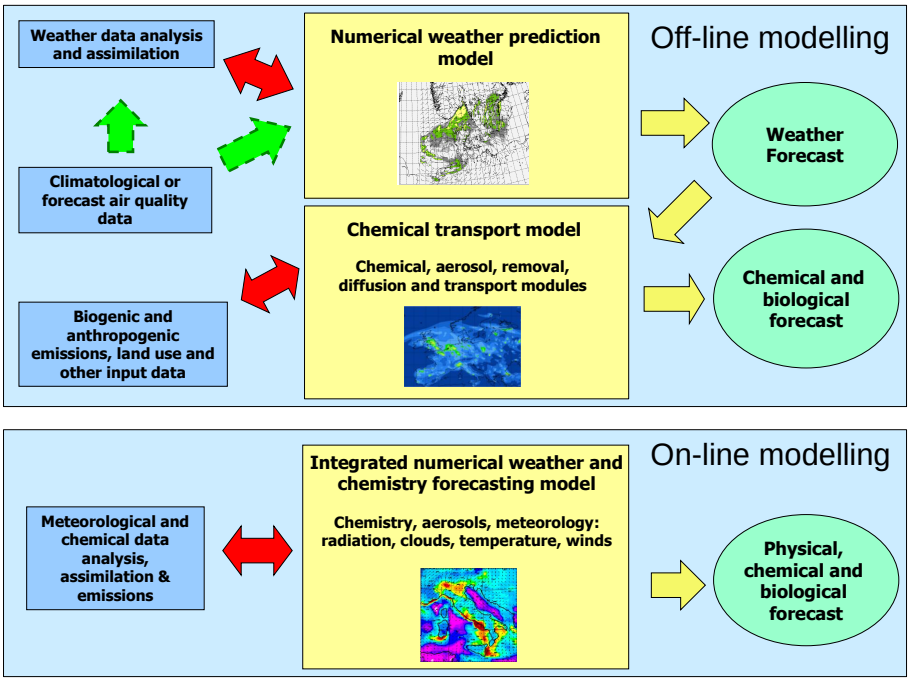

Fig. 1. Schematic diagram of the off-line and on-line coupled NWP and CTM modelling approaches for CWF.

On-line modelling systems (also called integrated or two-way interactive models) can be on-line access models, when meteorological data are available at each time-step (possibly via a model interface), or on-line integration of a CTM into a NWP model, where two-way feedbacks may be considered. We will use this latter definition for on-line coupled modelling.

The structure of on-line and off-line modelling systems has been schematically illustrated in Fig. 1.

The on-line integration of NWP or other meteorological models, with atmospheric aerosol and CTM allows all meteorological three-dimensional fields in CTMs at each time step to be used. It also facilitates the consideration of air-pollution feedbacks (e.g. those due to aerosols or greenhouse gases) on meteorological processes and climate forcing, and further on the chemical composition. Within the 18 CWF models considered here, only three models (Enviro-HIRLAM (Sect. 2.3.3), WRF-Chem (Sect. 2.3.18) and SKIRON/Dust (Sect. 2.3.16) in its new edition called ICLAMS-Integrated Community Limited Area Modeling System, Solomos et. al., 2011) are realised as on-line integrated models with two-way interactions. Previously, Zhang (2008) has reviewed the history and current status of the development and application of on-line coupled meteorology and chemistry models, with a focus on five representative models developed in the US including GATOR-GCMOM (Gas, Aerosol, Transport, Radiation, General Circulation, Mesoscale, and Ocean Model), WRF-Chem, CAM3 (Community Atmosphere Model v.3), MIRAGE (Model for Integrated Research on Atmospheric Global Exhanges), and Caltech unified general circulation model. An overview and description of existing on-line coupled chemistry-meteorology models in Europe was done by Baklanov et al. (2010b).

There are potential problems communicating between off-line coupled meteorological and CWF models. The advection schemes used in CWF models have to be improved for atmospheric chemistry transport models, as well as for NWP model, and it should be a high-priority task (e.g. Byun, 1999a, b; Baklanov et al., 2010b). For integrated atmospheric chemistry transport models, the requirements for advection schemes are even higher than for NWP models. They should be harmonised for all the scalars to maintain mass conservation and consistency. Thus, to achieve mass conservation, but at the same time maintain large time steps for the solution of dynamical equations, the models often include several advection schemes (such as semi-Lagrangian, Bott, SISL), which can be chosen in different combinations depending on the specific problem. The optimal way for online forecasting systems is to use the same conservative scheme for all the variables (e.g. for velocities, temperature, concentrations of chemicals, cloud water, humidity).

For offline atmospheric chemistry transport models, the choice of formulations and advection schemes is an independent and even more critically important problem (Byun, 1999a, b). Additionally, if NWP input data is used with non-conservative and unharmonized schemes (due to different schemes, grids, time steps in NWP and atmospheric chemistry transport models), the chemical part of offline

Table 1. Selected main characteristics of the 18 chemical weather forecasting (CWF) models considered in this study.

Model name	Coupling	Country and institution using it	CTM	NWP model	Type	Basic reference
ALADIN-CAMx	Off-line	ZAMG, Austria, Marcus Hirtl	CAMx	ALADIN-Austria	3-D Eulerian	http://www.cnrm.meteo.fr/aladin; http://www.camx.com
CAMx-AMWFG	Off-line	National and Kapodistrian University of Athens, Greece, George Kallos, Marina Astitha	CAMx	SKIRON/Dust	3-D Eulerian	http://forecast.uoa.gr/index.php; http://www.camx.com
ENVIRO-HIRLAM	On-line	DMI, Denmark, International HIRLAM team, Alexander Baklanov	Enviro	HIRLAM	3-D Eulerian	http://hirlam.org; www.hirlam.org/chemical
EURAD-RIU	Off-line	RIU, Cologne, Germany Hermann Jakobs, Hendrik Elbern, Michael Memmesheimer	EURAD	MM5	3-D Eulerian	http://www.eurad.uni-koeln.de
FARM	Off-line	ARIANET s.r.l. Italy, Giuseppe Calori, Camillo Silibello	FARM	RAMS	3-D Eulerian	http://www.aria-net.it/index_eng.php; http://www.minni.org/htm_farm2/Introduzione.htm
LOTOS-EUROS	Off-line	TNO/RIVM/PBL/KN, The Netherlands, P. J. H. Builtjes, M. Schaap, R. M. A. Timmermans	LOTOS-EUROS	ECMWF	3-D Eulerian	http://www.lotos-euros.nl/
MATCH	Off-line	SMHI, Sweden, Lennart Robertson, Thomas Klein	MATCH	ECMWF, HIRLAM	3-D Eulerian	http://www.smhi.se/sgn0106/if/meteorologi/match.htm; Robertson et al. (1999), Langner et al. (2005)
MM5-CAMx	Off-line	National and Kapodistrian University of Athens Aristotle University of Thessaloniki, Greece Zerefos Christos, Melas Dimitrios	CAMx	MM5	3-D Eulerian	http://www.mmm.ucar.edu/mm5; http://www.camx.com; http://lap.phys.auth.gr/gems.asp
MM5-CHIMERE	Off-line	Mesoscale Prediction Group in the Mesoscale and Microscale Meteorology Division, NCAR, Greece, Lia Fragkou (Model user)	CHIMERE	MM5	3-D Eulerian	http://www.mmm.ucar.edu/mm5; http://www.lmd.polytechnique.fr/chimere
MM5/WRF-CMAQ	Off-line	MM5-PSU/NCAR, Run operationally by the ESMG at Computer Science School of the Technical University of Madrid (UPM), Spain, Roberto San José WRF-NCAR/NCEP run routinely at the Centre for Atmospheric and Instrumentation Research (CAIR), University of Hertfordshire, UK, Ranjeet Sokhi	CMAQ	MM5/WRF	3-D Eulerian	http://www.mmm.ucar.edu/mm5; http://www.mmm.ucar.edu/wrf/users/; http://www.cmaq-model.org
MOCAGE	Off-line	Météo-France, Direction de la Production and Centre National de Recherches Météorologiques, France, Vincent-Henri Peuch (project leader)	MOCAGE	ARPEGE, ALADIN, ECMWF	3-D Eulerian	Dufour et al. (2004); http://www.prevair.org
NAME	Off-line	Atmospheric Dispersion Group, UK Paul Agnew	NAME	Met Office Unified Model	3-D Lagrangian	http://www.metoffice.gov.uk/research/modelling-systems/dispersion-model

Table 1. Continued.

Model name	Coupling	Country and institution using it	CTM	NWP model	Type	Basic reference
OPANA	Off-line	Environmental Software and Modelling Group, Computer Science School, Technical University of Madrid, LHTEE, AUT, NCAR/Pen, Spain, Roberto San José	OPANA	MEMO	3-D Eulerian	http://atmosfera.lma.fi.upm.es/equal/equal/show_long.htm http://artico.lma.fi.upm.es
RCG	Off-line	FU-Berlin, Institute for Meteorology Germany, Rainer Stern, Eberhard Reimer, Andreas Kerschbaumer	REM-CALGRID	GME	3-D Eulerian	http://www.trumf.de; Stern (2003)
SILAM	Off-line	Finnish Meteorological Institute, Finland, Mikhail Sofiev	SILAM	ECMWF, HIRLAM, WRF, AROME, ...	3-D Lagrangian, 3-D Eulerian	http://silam.fmi.fi
SKIRON/Dust	On-line	National and Kapodistrian University of Athens, Greece, George Kallos	SKIRON	Eta	3-D Eulerian	http://forecast.uoa.gr/dustinfo.php, Kallos et al. (2006), Spyrou et al. (2010)
THOR	Off-line	National Environmental Research Institute, Denmark, Jørgen Brandt	DEHM (UPM, OSPM)	Eta	3-D Eulerian (DEHM) 3-D Lagrangian (UPM)	http://thor.dmu.dk; http://www2.dmu.dk/atmosphericenvironment/thor/index.htm
WRF-Chem	On-line	NOAA 2008, Currently under research application in the ESMG-Computer Science School – Technical University of Madrid (UPM), Spain, Roberto San José	CHEM	WRF	3-D Eulerian	http://cprm.acd.ucar.edu/Models/WRF-Chem; http://ruc.noaa.gov/wrf/WG11/;

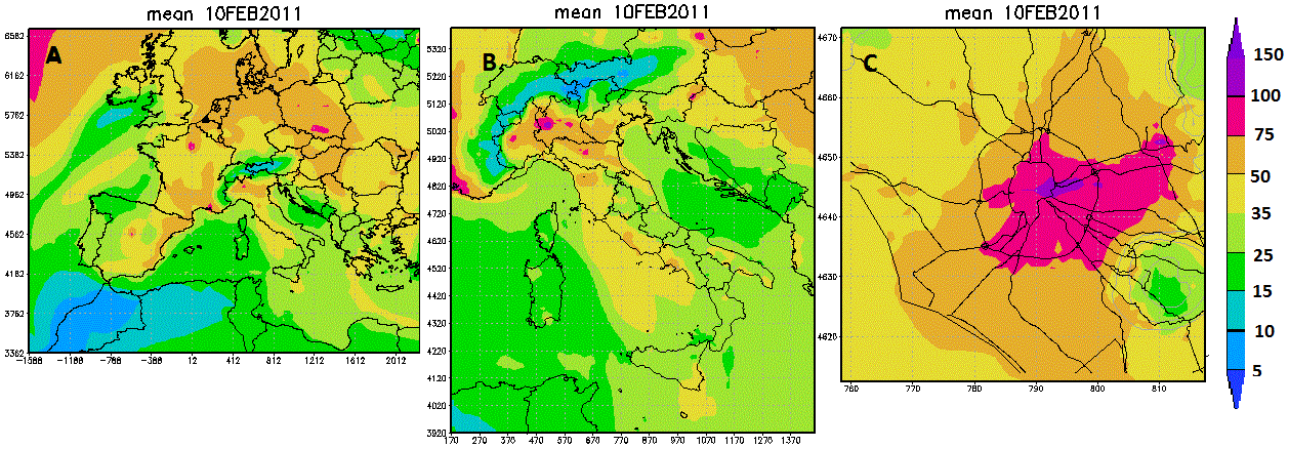

Fig. 2. The forecasted daily average concentrations of PM_{10} near the ground level on 10 February 2011, provided by the FARM model in **(a)** Europe, **(b)** Italy and **(c)** the urban area of Rome. The horizontal grid spacing in the forecasts of these three simulations are 48, 12 and 1 km, respectively. The scales on all the axes are in km, and the concentrations are in $\mu g\,m^{-3}$

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Table 2. Selected main characteristics of the numerical weather prediction models considered.

Model name	Hydrostatic/ nonhydrostatic	Vertical coordinate	Reference	Cloud microphysics	Convective parameterization scheme	PBL scheme	Global or Limited-area model
ALADIN	Hydrostatic	Pressure or Sigma-pressure hybrid	http://www.cnrm.meteo.fr/aladin/	Kessler (1969)	Bougeault (1985)	First order turbulence closure (Louis, 1979; Louis et al., 1982)	Limited-area
ECMWFIFS	Nonhydrostatic	Hybrid	http://www.ecmwf.int/research/ifsdocs/	Tiedke (1993)	Modified Tiedke (1989) scheme (Nordeng, 1994)	Modified Louis et al. (1982) K-theory scheme (Beljaars and Viterbo 1999)	Global
Eta	Nonhydrostatic	Eta	Mesinger et al. (1988), Janjić (1990, 1994)	Ferrier et al. (2002)	Betts-Miller-Janjić and Kain-Fritsch	Mellor-Yamada 2.5-order	Limited-area
GME	Hydrostatic	Sigma-pressure hybrid	Majewski et al. (2002)	Kessler-type scheme (Doms and Schättler, 1997)	Tiedtke (1989)	2nd order, Mellor and Yamada (1974)	Global
HIRLAM	Hydrostatic/ nonhydrostatic	Sigma-pressure hybrid	http://hirlam.org	STRACO (Soft Transition Condensation)	STRACO (modified Kuo scheme), Rasch and Kristjánsson (1998), Kain-Fritsch	Cuxart Bougeault Lacarrere, order 1.5 TKE scheme	Limited-area
MEMO	Nonhydrostatic	Sigma	Kunz and Moussiopoulos (1995), Moussiopoulos et al. (1997)	No moist processes	No moist processes	K-theory	Limited-area
MM5	Nonhydrostatic	Sigma	Dudhia (1993), Grell et al. (1995)	Various possible schemes	Various possible schemes	Various possible schemes	Limited-area
Unified Model	Nonhydrostatic	Height	Cullen et al. (1997), Davies et al. (2005)	Wilson and Ballard extended by Forbes	Gregory and Rowntree (1990)	Lock et al. (2000)	Limited-area or global
WRF	Nonhydrostatic	Sigma or sigma-pressure hybrid	Janic et al. (2001), Janic (2003), Skamarock et al. (2005)	Various possible schemes	Various possible schemes	Level 2.5 Mellor and Yamada Janic, or non local YSU scheme	Limited-area
RAMS	Nonhydrostatic	Height, sigma or eta	Pielke et al. (1992), Cotton et al. (2003)	Various possible schemes	Various possible schemes	Various possible schemes	Limited-area or global

atmospheric chemistry transport models can produce unrealistic forecasts. For online coupling, it is not a problem if one uses the same mass-conservative scheme for chemistry, cloud water and humidity. The wide variety of existing modelling systems has led to a number of approaches and methods implemented in interface modules. Tasks performed by interfaces are minimised in some coupled systems (as online models do not need interfaces per se); these rely on surface fluxes, and turbulence and dispersion parameters (such as eddy viscosity) that are provided by the meteorological drivers.

Other modelling systems use interface modules that implement surface and boundary-layer parameterisations to estimate dispersion parameters. In some cases, interfaces are used to enhance the resolution of local physiographic data, and possibly introduce advanced parameterisations (e.g. those for urbanisation). Moreover, interface modules can include the evaluation of emissions of species that can be strongly influenced by meteorology, such as biogenic Volatile Organic Compounds (VOC), windblown dust, sea salt, and pollen.

2.3 Overview of the CWF modelling systems

The 18 models discussed in this article are now briefly introduced, presented in alphabetical order by their acronyms. A summary of selected main characteristics of these models appear in Table 1. The aim of this section is to give a quick overview of the models, before a more detailed and systematic examination of their properties. The European countries of the model users are listed in the titles. In some cases, these countries may differ from the countries where these models were originally developed.

Selected examples on operational forecast products have been illustrated in Fig. 2a–c. The European- and Italian-scale forecasts were computed using the same two-way nested simulations, whereas forecasts for Rome were performed independently using the Italian-scale simulation results as

Table 3. Brief characterizations of the main physical and chemical components of the CWF models.

Model name	Advection and convection	Diffusion	Dry deposition	Wet deposition	Chemistry package	Aerosol package
ALADIN-CAMx	Eulerian continuity equation closed by K-theory equations in flux form. The advection scheme of Bott (1989) is used	Horizontal diffusion: Smagorinsky approach. Vertical diffusion: Louis (1979) approach (uses the Richardson number and the mixing length)	Physical: separate resistance models for gases and aerosols. Numerical: deposition velocity as surface boundary condition for vertical diffusion	Physical: seperate scavenging models for gases and aerosols. Numerical: uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size	SAPRC-99: 114 species, 217 reactions	The CW scheme is used for aerosol chemitry. It divides the size distribution into two static modes (coarse and fine). Primary species are modeled as fine and/or coarse particles, while all secondary species are modeled as fine particles only
CAMx-AMWFG	Eulerian continuity equation closed by K-theory. The advection solver used is The Area Preserving Flux-Form advection solver of Bott (1989)	Horizontal diffusion based on Smagorinsky approach. Vertical diffusion coefficients supplied via input file (from the meteorological model)	Separate resistance models for gases and aerosols. Numerical: deposition velocity as surface boundary condition for vertical diffusion	Separate scavenging models for gases and aerosols. Numerical: uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size	Carbon Bond (CB-IV)	RADM aqueous chemistry scheme, ISORROPIA gas/aerosol partitioning scheme, SOAP scheme for SOA formation, both Coarse/Fine scheme and Multi sectional approach. Options for two variable (coarse/fine) bulk scheme and fixed sectional scheme (sections by user choice) with all microphysics. 16 aersosol chemical species. (Sulfate, Nitrate, Ammonium, Water, Anthropogenic SOA, Biogenic SOA, Polymerized anthropogenic SOA, Polymerized biogenic SOA, Sodium, Chloride, Primary Organic Aerosol, Primary Elemental Carbon, Fine Other Primary, Fine Crustal, Coarse Other Primary, Coarse Crustal)
Enviro-HIRLAM	Several possibilities: globally and locally-conservative schemes for advection (Bott, 1989; Kaas, 2008). STRACO convection and Tiedtke mass-flux schemes to convect aerosols and gases. Eulerian continuity equation closed by K-theory	Vertical diffusion by native TKE-l scheme (Cuxart, 2000). Horizontal diffusion by the native extra scalar system. Improved parameterisation for urban boundary layer and mixing height	Separate resistance models for gases and aerosols. Three regimes gravitaton settling parameterisations for different size aerosols	Separate 3-D scavenging models for gases and aerosols, and for rain-out and wash-out with particle size-dependent parameterisations (Baklanov and Sørensen, 2001)	Four mechanisms for gas-phase chemistry can be used: NWP-Chem (default scheme), RADM2, RACM and an extended version (includes isoprene and DMS chemistry) of CBMZ	Aerosol module comprises thermodynamic equilibrium model NWP-Chem-Liquid and an aerosol dynamics model (Korsholm et al., 2008). 4 aerosol modules: modal CAC (default, Gross and Baklanov, 2004) and MADE (Ackermann et al., 1998), and sectional MOSAIC (Zaveri et al., 2007) and SALSA (on test phase, Kokkola et al., 2008)
EURAD-RIU	fourth order Bott scheme (Bott, 1989)	Vertical mixing based on scaling regimes (Holtslag et al., 1990)	Deposition velocity based on landuse type and season. Revised parameterization by Zhang et al. (2003)	Gas-phase: Henrys law equilibria for all prognostic species. Aerosol phase (Binkovski, 1999): the accumulation mode particles form cloud condensation nuclei and are 100 % absorbed into the cloud water. The Aitken mode forms interstitual aerosol which is scavenged by cloud droplets. The wet removal of aerosol is proportional to the wet removal of sulfate	RADM2 (Stockwell et al., 1990), RACM (Stockwell et al., 1997), Euro-RADM (Stockwell and Kley, 1994) – The Euro-RADM chemical mechanism was developed to model European atmospheric chemistry. It is based upon the Regional Acid Deposition Model mechanism, version 2 (RADM2)	The aerosol dynamics model MADE (Ackermann et al., 1998), SORGAM (Schell et al., 2001)
FARM	Horizontal: Blackman cubic polynomials (Yamartino, 1993). Vertical: hybrid semi-implicit Crank-Nicolson/fully implicit scheme (Yamartino et al., 1992)	Horizontal diffusion based on Smagorinsky approach or stability dependent parameterizations. Different vertical diffusion parameterizations based on PBL-scaling. K_h and K_z evaluated by SURFPRO pre-processor	Deposition velocity (from SURFPRO pre-processor) depending on land type, season, surface meteorology, surface wetness, by means of a big leaf resistance model after Walcek et al. (1986) and Wesely (1989)	Precipitation scavenging based on EMEP (2003)	Two mechanisms implemented through KPP chemical pre-processor (Sandu et al., 1995): an updated version of the chemical mechanism implemented in the EMEP Lagrangian Acid Deposition Model (Hov et al., 1988) including the treatment of Persistent Organic Polluttants (POPs) and mercury and SAPRC-99 (Carter, 2000). Photolysis reaction rates estimated either using simple look-up tables or an on-line version of the Tropospheric Ultraviolet-Visible Model (TUV, Madronich, 1987). Cloud effects on actinic flux considered	CMAQ aero3 module to includes aerosol processes: modal scheme with three modes and all microphysics. ISORROPIA and SORGAM models to include aerosol thermodynamics/ partitioning respectiverly for inorganic and organic species

Table 3. Continued.

Model name	Advection and convection	Diffusion	Dry deposition	Wet deposition	Chemistry package	Aerosol package
LOTOS-EUROS	Advection following Walcek (2000). No explicit convection, vertical grid follows pbl from meteorology	Vertical turbulent mixing formulation uses K-diffusion	Resistance approach	Scavenging rates depending on Henry's law constants for gases and following Scott (1978) for particles	Updated CBM-4 with Carter's 1-product isoprene scheme: homo- and heterogenous conversion of NO_2 to HNO_3. 28 species and 66 reactions, including 12 photolytic reactions	Bulk scheme with possibility for several size ranges. ISORROPIA, MARS or EQSAM options for calcluating equilibrium between gas and particle phase sulfate, nitrate, ammonium, water
MATCH	Modeled using a Bott-type advection scheme (Bott, 1989). Up to forth order schems are implemented in the horozontal and up to second order in the vertical	Parameterized using three primary parameters: the surface friction velocity, the surface sensible heat flux and the boundary layer height	Modelled using a resistance approach. Deposition schemes with different degrees of sophistication are available	Assumed to be proportional to the precipitation intensity using species-specific scavenging coefficients. For particles, several different schemes are avalable	Extended EMEP MSC-W model chemistry (Simpson et al., 1993). Aqueous-phase oxidation is implemented following Berge (1993). The formation of ammonium sulfate and -nitrate is modelled following Hov et al. (1994). 110 thermal, 28 photo-chemical, 2 aqueous-phase, 5 aerosol reactions and 4 gas-phase aqueous-phase and aerosol equlibria between 60 chemical components	Bulk scheme with several non-interacting size ranges. Equilibrium between particle and gas phase
MM5-CAMx	MM5: vertical advection of moisture and temperature are resolved by applying linear interpolation methods. Convection in cumulus clouds is parameterized with the Kain-Fritsch 2 parameterization. CAMx: Eulerian continuity equation closed by K-theory	MM5: MRF Planetary Boundary-Layer (PBL) scheme. CAMx: horizontal diffusion based on Smagorinsky approach. Vertical diffusion coefficients supplied via input file (from the meteor-ological model)	CAMx: physical: separate resistance models for gases and aerosols. Numerical: deposition velocity as surface boundary condition for vertical diffusion	Separate scavenging models for gases and aerosols. Numerical: uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size	CAMx: Carbon Bond (CB-IV)	RADM aqueous chemistry algorithm, ISORROPIA inorganic aerosol thermodynamics/partitioning, SOAP scheme for SOA formation. Options for two variable (coarse/fine) bulk scheme and fixed sectional scheme (sections by user choice) with all microphysics. 16 aersosol chemical species (sulfate, nitrate, ammonium, water, anthropogenic SOA, biogenic SOA, polymerized anthropogenic SOA, polymerized biogenic SOA, sodium, chloride, primary organic aerosol, primary elemental carbon, fine other primary, fine crustal, coarse other primary, coarse crustal)
MM5-CHIMERE	MM5: based on a finite difference formulation of the time-dependent Navier-Stokes equations. CHIMERE: 3 advection schemes: the Parabolic Piecewise Method, the Godunov scheme and the simple upwind first-order scheme	MM5: bulk PBL, high resolution Blackadar PBL, Burk. Thompson PBL, Eta PBL, MRF PBL, Gayno-Seaman PBL, Pleim-Chang PBL. CHIMERE: vertical turbulent mixing takes place only in the boundary-layer. The formulation uses K-diffusion, without counter-gradient term	CHIMERE: considered for model gas species and is parameterized as a downward flux out of the lowest model layer. The deposition velocity is described through a resistance analogy (Wesely 1989)	MM5: nonconvective precipitation scheme, warm rain, simple ice, mixed-phase, Goddard microphysics, Reisner graupel, Schultz microphysics. CHIMERE: follows the scheme proposed bu Loosmore (2004)	CHIMERE: offers the option to include different gas phase chemical mechanisms. MELCHIOR1 (Lattuati, 1997): more than 300 reactions of 80 gaseous species. The hydro-carbon degradation is fairly similar to the EMEP gas phase mechanism. MELCHIOR2: 44 species and about 120 reactions is derived from MELCHIOR (Derognat, 2003), following the concept of chemical operators (Carter, 1990)	Sectional with 6 size bins (each bin internally mixed). All microphysical processes included. 7 species (primary particle material, nitrate, sulfate, ammonium, biogenic secondary organic aerosol (SOA), anthropogenic SOA and water)
MM5/WRF-CMAQ	Several possibilities. Normally Global-mass conserving scheme. Vertical difussion is mainly done with the Asymmetric Convective model (ACM2) for MM5 and the Yonsei University (YSU) PBL parameteriza-tion for WRF	PBL scheme and diffussion (MRF PBL); surface scheme: Noah Land Surface Scheme and Monin Obukhov surface layer scheme	Physical: separate resistance models for gases and aerosols. Numerical: deposition velocity as surface boundary condition for vertical diffusion	Physical: seperate scavenging models for gases and aerosols. Numerical: uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size	CB04 and CB05; Also RADM chemistry. It includes cloud and aerosol chemistry	Modal scheme with three modes and all microphysics. Aerosol species: elemental and organic carbon, dust, and other species not further specified. Secondary species considered are sulfate, nitrate, ammonium, water, and secondary organics from precursors of anthropogenic and biogenic origin. Possibilities to run the aerosol MADRID scheme

Table 3. Continued.

Model name	Advection and convection	Diffusion	Dry deposition	Wet deposition	Chemistry package	Aerosol package
MOCAGE	Advection is based upon the semi-lagrangian scheme of (Williamson and Rasch, 1989) with global mass conversation imposed, while convection is parameterised using (Bechtold et al., 2001). See details and evaluation in (Josse et al., 2004)	K-diffusion approach, based upon (Louis, 1979)	Resistance approach: for gases, it is based upon (Wesely, 1989) with refinements for stomatal resistance (Michou et al., 2004); for aerosol, the approach is described in (Nho-Kim et al., 2004)	For scavenging by large-scale precipitation and below convective cloud, the parameterisation is based upon (Giorgi and Chameides, 1986), with a special treatment for snow flakes. For scavenging within convective clouds, it is done within the convective paramaterisation as described in (Mari et al., 2000)	Several options are available. In the operationnal version, the scheme is a merge from the schemes RACM (Stockwell et al., 1997) and REPROBUS (Lefevre et al., 1994), thus offering a comprehensive representation of both tropospheric and stratospheric chemistry. The scheme comprises 118 species for a total of over 300 homogeneous and heterogeneous chemistry reactions	Aerosol is described using a bulk approach with size bins (typically 5 to 20 bins per species). Evaluation is currently available for black carbon (Nho-Kim et al., 2005), dust (Martet et al., 2009) and sulfate (Ménégoz et al., 2009)
NAME	NAME does not have its own convection scheme. It obtains advection fields from the UK Met. Office's numerical weather prediction model, the Unified Model (Cullen, 1993)	Modelled using random walk techniques. Parameterised profiles of turbulence parameters such as velocity variances and Lagrangian time scales are employed. Normally a Gaussian scheme is used. However for convective conditions a skewed turbulence scheme is available	Uses a resistance analogy approach and sedimentation of heavy particles is also included	Based on parameterised scavenging coefficients and differs according to precipitation type (convective, dynamic)	Based on the scheme for the STOCHEM model. 40 advected tracers + 18 non-advected 140 reactions + 23 photolytic reactions 16 emitted species	A mass based scheme incorporating sulphate, nitrate, ammonium, and secondary organic aerosols. Additional scheme exist for sea salt (2 size bias) and mineral dust (6 size bias)
OPANA	Piecewise parabolic method (PPM) which is a monotonic scheme with geometric non-linear adjustments to the parabolic concentration distributions	Two convective boundary-layer schemes: Blackadar and ACM. Local diffusion, vertically continuous integration, smooth transition from stable to convective and faster matrix solver. Updated eddy diffusion scheme	Chemical gases: Wesely (1989). Aerosol chemistry: Binkowski and Shankar (1995) approach. These schemes are based on the resistance approach which assumes a canopy, aerodynamical and bulk resistance	Physical: seperate scavenging models for gases and aerosols. Numerical: uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size	The CBM-IV chemical mechanism in short and long modes are included in the system. The RADM model and the SAPRC-99 chemical scheme are also included. These schemes simulate the chemical reactions in the atmosfere for organic and also inorganic reactions	Modal scheme with three modes and all microphysics. Aerosol species: elemental and organic carbon, dust, and other species not further specified. Secondary species considered are sulfate, nitrate, ammonium, water, and secondary organics from precursors of anthropogenic and biogenic origin. Possibilitirs to run the aerosol MADRID scheme
RCG	Walcek (2000). The number of steps within the advection scheme is chosen such that the courant restriction is fulfilled	Vertical turbulent mixing formulation uses K-diffusion. Stable and convective boundary-layer diffusion coefficients based on PBL scaling regimes. In addition: mixing by use of time and space dependent coordinate based on mixing height	Resistance approach (Erisman et al., 1994). Gases: function of species dependent Henry constant and precipitation rate. Particles: below-cloud simplescavenging coefficient approach with identical coefficients for all particles	Gases: function of species dependent Henry constant and precipitation rate. Particles: below-cloud simplescavenging coefficient approach with identical coefficients for all particles	Gas phase: updated CBM-4 with Carter's 1-product isophere scheme: homo- and heterogenous conversion of NO_2 to HNO_3, aqueous phase conversion of SO_2 to H_2SO_4 through oxidation by H_2O_2 and O_3, equilibrium concentration fot SO_2, H_2O_2 and O_3 from Henry constants	ISORROPIA. Bulk equilibrium scheme. species: mineral coarse between 2.5 µm and 10 µm, primary aerosol smaller than 2.5 µm, primary elemental carbon, primary organic carbon, secondary organic aerosols, sulfate, nitrate, ammonium, sea-salt
SILAM	Lagrangian kernel uses the iterative advection of Eerola (1990). Eulerian kernel is built on the basis of Galperin (2000)	Lagrangian: assumes the well-mixed PBL and fixed random-walk parameters in the free troposphere. Exchange between the PBL and troposphere takes place due to variation of the PBL height. Eulerian: follows the K-closure with approach of Gernikhovich et al. (2004) and Sofiev et al. (2010a) for K_z evaluation	Varies for different chemical schemes but generally is based on resistive analogy with certain simplifications or extensions. Aerosols: both gravitational and diffusional paths are considered with the sedimentation treated via Stokes terminal velocity	Follows the scavenging coefficient approach, distinguishing between the in- and sub-cloud, as well as rain-, snow-types of scavenging	1. Acid basic transformations 2. CB-4 3. Sox module 4. Radioactive decay of up to ∼ 500 nuclides 5. Toxic species 6. Aerosols 7. Natural birch pollen 8. Sea salt 9. Passive tracer	Two schemes: bulk and ADB (Aerosol Dynamics Basic – research mode only). Both schemes use the user-defined set of bins. Bulk scheme allows treatment of any chemically inert aerosol. ADB scheme distinguishes between SIA, sea salt, dust, primary PM – in soluble and insoluble phases
SKIRON/Dust	Horizontal: the Eta/NCEP model scheme for advection of a passive substance (Janjić, 1997). Vertical: the scheme of Van Leer (1977)	A 2nd order diffusion scheme is used for lateral diffusion by utilizing the Smagorinsky-type horizontal diffusion coefficient modified by the presence of the model turbulent kinetic energy term (Janjić, 1990)	Surface deposition of particles occurs via diffusion, impaction, and/or gravitational settling using the resistance approach of Slinn and Slinn (1980)	Wet deposition of particles occur above and below the clouds as described by Seinfield and Pandis (1998)	No chemistry	Bulk scheme for dust

Table 3. Continued.

Model name	Advection and convection	Diffusion	Dry deposition	Wet deposition	Chemistry package	Aerosol package
THOR	Time integration for the advection term is performed with a predictor-corrector scheme with several correctors (Zlatev, 1995)	K-theory, constant K in horizontal, and vertical K_z based on Monin-Obukhov similarity theory for the surface layer, extended to the whole mixing layer	DEHM: velocities of the species are based on the resistance method	DEHM: parameterised by a simple scavenging ratio formulation with different in-cloud and below-cloud scavenging	A chemical scheme similar to the EMEP scheme with 60 species and 120 chemical reactions is included in the model	DEHM: Bulk scheme. Three of the species are primary particulates (PM_{25}, PM_{10}, TSP). Furthermore sea salt is implemented in the model
WRF-Chem	Several possibilities. Normally Global-mass conserving scheme. Vertical difussion is mainly done with the Yonsei University PBL parameterization	PBL scheme with the Yonsei University parameterization	Physical: separate resistance models for gases and aerosols. Numerical: deposition velocity as surface boundary condition for vertical diffusion	Physical: separate scavenging models for gases and aerosols. Numerical: uptake as a function of rainfall rate, cloud water content, gas solubility and diffusivity, PM size	CB05, CBMZ and RADM chemical schemes	Option for sectional (MOSAIC, 8 bins) and modal (MADE) aerosol models

boundary conditions. The date of the forecasts (10 February 2011) corresponds to a severe air-quality episode, during which concentrations of PM_{10} (Particulate Matter $<10\,\mu m$) in the European, and Italian and Roman domains were substantial.

2.3.1 ALADIN-CAMx (Austria)

The chemical weather model for Austria, AQA (Air-Quality model for Austria), consists of the meteorological model ALADIN-Austria (Sect. 3.2.5) and the chemical transport model CAMx (Comprehensive Air-Quality Model with extensions; http://www.camx.com). The two models are coupled off-line. The modelling system ALADIN-CAMx was implemented for the first time in Baumann-Stanzer et al. (2005). The forecasts, which are done in cooperation with the University of Natural Resources and Applied Life Sciences in Vienna (BOKU), are supported by the regional governments in Austria.

The SAPRC99 (Statewide Air Pollution Research Center in Riverside, California, Sect. 4.3.7) gas-phase photochemistry module (Carter, 2000) used in the operational AQA forecasts considers 76 different species and 217 reactions. The model system generally uses European Monitoring and Evaluation Programme (EMEP; http://www.emep.int) emissions for Europe. For the countries Austria, Czech Republic, Slovakia and Hungary, the original 50×50 km data are downscaled to 5×5 km based on an inventory from 1995. The EMEP data for 2005 (Vestreng et al., 2006) was used during summer 2007. In addition, a new highly resolved emission inventory for the City of Vienna (Orthofer et al., 2005) is used. Before 2008, terpene and isoprene emissions were calculated according to Guenther et al. (1993), and biogenic NO and NO_2 emissions were calculated according to Williams et al. (1987) and Stohl et al. (1996). For the operational chemical weather forecasts in 2009, these methods were replaced by the BEIS3 (Biogenic Emission Inventory System) mechanism, which is implemented in the emission model SMOKE (Sparse Matrix Operator Kernel Emissions model).

Monthly average (1991–2001) concentrations of the different species are used as boundary conditions for the coarse grid. The concentrations were obtained from model calculations (Krüger et al., 2008), which were conducted for the EU-project CECILIA (Central and Eastern Europe Climate Change Impact and Vulnerability Assessment, http://www.cecilia-eu.org). Forecasts of total column ozone are provided by the ECMWF IFS (Integrated Forecast System) model (Sect. 3.2.1).

2.3.2 CAMx-AMWFG (Greece)

The AM&WFG (Atmospheric Modelling and Weather Forecasting Group, National and Kapodistrian University of Athens, Greece) developed the chemical weather forecasting system CAMx-AMWFG, which is based on the CAMx photochemical model. The system utilizes the SKIRON/Dust modelling system (Sect. 2.3.16) meteorological fields in order to prepare long-range transport of gases and particulate matter for Europe and the Mediterranean Region. The CAMx model was developed for regional-scale modelling of ozone and other pollutants (ENVIRON, 1997, 2006). Products from this model are 48-h operational forecasts of O_3, NO_2, SO_2, and particulate sulfate (PSO_4) fields for the Mediterranean region and Europe every hour produced once per day (http://forecast.uoa.gr/camxindx.php). Additionally, CAMx-AMWFG can provide the concentration and deposition for sodium and chloride (from sea-salt production), sulfate produced on dust (DSO_4), and nitrate produced on dust (DNO_3).

2.3.3 Enviro-HIRLAM (Denmark and others)

Enviro-HIRLAM (Environment – HIgh Resolution Limited Area Model) is an on-line coupled NWP (Sect. 3.2.4) and CTM model for research and forecasting of both meteorological and chemical weather. The modelling system was developed by DMI (Danish Meteorological Institute) with

other collaborators (Chenevez et al., 2004; Baklanov et al., 2008b; Korsholm et al., 2008) and included by the European HIRLAM consortium as the baseline system in the HIRLAM Chemical Branch (www.hirlam.org/chemical).

To make the model suitable for CWF in urban areas, the meteorological part is improved by implementation of urban sublayer modules and parameterisations. The aerosol module in Enviro-HIRLAM has two parts: (i) a thermodynamic equilibrium model (NWP-Chem-Liquid) and (ii) the aerosol dynamics model CAC (tropospheric Chemistry Aerosol Cloud model). Parameterisations of aerosol feedback mechanisms in the Enviro-HIRLAM model are described in Korsholm et al. (2008) and Korsholm (2009).

Users have the option to choose one of several chemical mechanisms: RADM2 (Sect. 4.3.6), RACM (Sect. 4.3.6) or the newly developed, economical NWP-Chem. On-line Enviro-HIRLAM is used at DMI for operational pollen forecasting. The DMI operational system also includes the off-line version (the so-called CAC system), which is used operationally for CWF (e.g. in GEMS) and the Lagrangian model DERMA (Danish Emergency Response Model of the Atmosphere; Sørensen et al., 2007) for emergency preparedness modelling.

2.3.4 EURAD-RIU (Germany)

The EURAD model (European Air Pollution and Dispersion model) is an chemical weather forecast model system for research and assessment. The model system was developed at the Rhenish Institute for Environmental Research (RIU) at the University of Cologne, Germany. The EURAD Air Quality Forecast System consists of three major components: MM5 (Sect. 3.2.8) to predict the needed meteorological variables, the EURAD Emission Module (EEM) to calculate the temporal and spatial distribution of the emission rates of the major pollutants, and the EURAD Chemistry Transport Model (EURAD-CTM) to predict the concentrations and deposition of the main atmospheric pollutants.

For the initial and boundary conditions, the NCEP GFS (National Centers for Environmental Prediction, Global Forecast System) is interpolated onto the grids of the nested MM5 domains. The nesting enables consistent modelling from local to continental scales. Geographical information (e.g. orography, land-use type) is taken from the United Sates Geological Survey (USGS) database.

EURAD uses the RADM2 and its successor RACM for computing the chemical processes and MADE (Modal Aerosol-Dynamics model for EURAD) for computing aerosol processes. RADM2 contains 63 reactive species treated in 158 chemical reactions. There is an option to run the code with the more sophisticated RACM chemistry. Detailed aqueous-phase chemistry is incorporated, as well. The horizontal and vertical transport is performed by the fourth-order Bott advection scheme, and vertical mixing of the species is treated by an implicit vertical diffusion scheme. The sink at the lower boundary of the model is treated by wet and dry deposition parameterization. The major driver for wet deposition is the predicted precipitation. The dry deposition is calculated via the deposition velocity for each species, which depends upon the properties of the particle, the meteorological conditions and the land-use type.

The daily output of meteorological and atmospheric constituents covers Europe, Central Europe and the German States of North Rhine-Westfalia, Lower Saxony and Bavaria. These products are published on the EURAD website (http://www.riu.uni-koeln.de).

2.3.5 FARM (Italy)

FARM (Flexible Air-quality Regional Model) was originally derived from STEM-II (Sulfur Transport and dEposition Model; Carmichael et al., 1998) and was later developed as an independent project by ARIANET s.r.l. (http://www.aria-net.it). The model development is presently supported by ENEA (Ente per le Nuove tecnologie, l'Energia e l'Ambiente; www.enea.it) within the national project MINNI (Modello Integrato Nazionale a supporto della Negoziazione Internazionale sui temi dell'inquinamento atmosferico; www.minni.org) funded by the Italian Ministry of Environment. A short model presentation is available at http://www.aria-net.it/front/ENG/codes/files/10.pdf, a more detailed on-line description (in Italian) can be found at http://www.minni.org/sistema/sistema-modellistico-atmosferico/modulo-chimico, and a comprehensive user's guide is provided on request. Recent applications are documented in Gariazzo et al. (2007), Silibello et al. (2008) and Calori et al. (2008).

SAPRC-99 (Carter, 2000) and an updated version of the chemical mechanism implemented in the EMEP Lagrangian Acid Deposition Model (Hov et al., 1988), including the treatment of Persistent Organic Pollutants (POPs) and mercury, and gas-phase chemical mechanisms have been implemented into the model using KPP chemical pre-processor (KPP, Kinetic Pre-Processor: Damian et al., 2002; Sandu et al., 2003; Daescu et al., 2003). The integration of the chemical reactions is performed by means of the the following methods included in KPP: Rosenbrock (Sandu et al., 2003) and LSODE (the Livermore Solver for Differential Equations; Radhakrishnan and Hindmarsh, 1993). Photolysis reaction rates appearing in the mechanism can be estimated either using simple look-up tables or an on-line version of the Tropospheric Ultraviolet-Visible Model (TUV, Madronich, 1987). SAPRC99 is coupled with the CMAQ (Community Multiscale Air-Quality model) aero3 (3rd generation aerosol) module to include aerosol processes. In the presence of a cloud layer, a simplified aqueous-phase mechanism includes sulfate production in clouds.

FARM runs operationally at ARIANET coupled with the meteorological model RAMS (Regional Atmospheric Model System; Cotton et al., 2003, Sect. 3.2.10) to

produce national-scale chemical weather forecasts (http://www.aria-net.eu/QualeAria). The model is run by some Italian Regional Environmental Protection Agencies (ARPA) to produce chemical weather analyses and forecasts. In particular, ARPA Lazio runs the model driven by RAMS to produce urban chemical weather forecasts for Rome (Finardi et al., 2009) available at http://www.arpalazio.net/main/aria/sci/previsioni/pm10.php; ARPA Piemonte together with Novara and Torino Provinces use the model driven by COSMO I7 (Italian implementation of the LM model, formerly known as LAMI, Limited Area Model Italy, http://www.arpa.emr.it/sim/?mappe_numeriche) to forecast air quality over Torino City (Finardi et al., 2008) and Novara Province (Pittini et al., 2007), with results browsable at http://www.provincia.torino.it/ambiente/inquinamento/aria/qualita/ipqa/index and at http://www.provincia.novara.it/arianova/WEB/index.html; and ARPA Lombardia uses FARM and a meteorological analysis using the ECMWF as a background field to compute near-real-time chemical weather analyses (http://ita.arpalombardia.it/ITA/qaria/doc_DistribSpazialeCalcolata.asp).

2.3.6 LOTOS-EUROS (The Netherlands)

Several models have been developed in The Netherlands. Netherlands Organisation for Applied Scientific Research (TNO) developed LOTOS (Builtjes, 1992; Schaap et al., 2004) and the National Institute for Public Health and the Environment (RIVM) developed EUROS (de Leeuw and van Rheineck Leyssius, 1990; Matthijsen et al., 2002). During 2004, the two models were unified, resulting in the LOTOS-EUROS model version 1.0 (LOng Term Ozone Simulation – EURopean Operational Smog model, Schaap et al., 2008; http://www.lotos-euros.nl). The model can be used to model the fate of pollutants such as photo-oxidants, aerosols, heavy metals and POPs over Europe.

The model is used operationally to forecast air pollution over Europe and the Netherlands, driven by the meteorology from ECMWF IFS (Sect. 3.2.1). The model is used to perform 72-h European forecasts (twice daily) at 30- and 15-km horizontal grid spacing and a smaller domain over the Netherlands at a 15-km horizontal grid spacing, including data assimilation of ozone measurements (van Loon et al., 2004). PM_{10} forecasts are produced including a bias correction (Manders et al., 2009).

2.3.7 MATCH (Sweden)

The Multi-scale Atmospheric Transport and Chemistry (MATCH) model is a three-dimensional, Eulerian model developed at SMHI (Swedish Meteorological and Hydrological Institute). It is used in a range of applications from urban-scale studies with grid spacings on the order of a km or smaller (e.g. Gidhagen et al., 2005) to regional and continental-scale studies on eutrophic deposition and photochemistry (e.g. Langner et al., 2005; Engardt et al., 2005; Andersson et al., 2007). MATCH is used for air-quality assessment in Sweden and the Baltic Sea region and for forecasts of radioactivity in case of nuclear emergencies in Europe (Langner et al., 1998).

MATCH includes modules describing emissions, advection, turbulent mixing and dry and wet deposition. Depending on the application, specific modules describing chemistry or aerosol dynamics can be added to the basic transport model. The MATCH design has flexible horizontal and vertical resolution and allows for an arbitrary number of chemical compounds. The advection scheme (Bott, 1989) is fourth-order in the horizontal and second-order in the vertical. A complete description of the transport model can be found in Robertson et al. (1999) and in the on-line documentation (http://www.smhi.se/sgn0106/if/meteorologi/match.htm). Details on the photochemistry version of MATCH can be found in Andersson et al. (2007) and van Loon et al. (2007). Emissions used for the runs are based on EMEP2003.

The current MATCH operational system for CWF consists of two components, driven by HIRLAM and ECMWF meteorology, respectively. The MATCH-HIRLAM component uses meteorological data provided by the HIRLAM NWP model (Sect. 3.2.4) that is operational at SMHI. MATCH-HIRLAM is primarily targeted to ozone and is run once a day. A run consists of a hindcast of the previous day and forecasts for the present day and the next day. The model grid comprises nearly all of Europe with a horizontal grid spacing of 44 km (http://www.airviro.smhi.se/MAQS). MATCH-ECMWF is a part of the GEMS/MACC regional cluster and uses ECMWF IFS (Sect. 3.2.1) meteorology. It is currently operated with horizontal grid spacing of 0.5 and 0.2° (since 2011) and provides 72-h forecasts of such quantities as O_3, NO, NO_2, CO, SO_2, $PM_{2.5}$, PM_{10}, visibility and aerosol optical depth (AOD) (http://gems.ecmwf.int/).

2.3.8 MM5-CAMx (Greece)

MM5-CAMx is the combination of the limited-area, non-hydrostatic, terrain following and sigma-coordinate meteorological model MM5 (Sect. 3.2.8) coupled off-line with the three-dimensional Eulerian photochemical transport model CAMx. The forecast system performs a 72-h forecast of daily mean and daily maximum O_3, NO, NO_2, CO, SO_2, and PM_{10} concentrations on three domains: two on the regional scale (Europe, Balkan Peninsula) and one on an urban scale (Athens) (http://lap.phys.auth.gr/gems.asp).

2.3.9 MM5-CHIMERE (France and Portugal)

MM5-CHIMERE consists of two models: the PSU/NCAR (National Center for Atmospheric Research) Mesoscale Model MM5 (Sect. 3.2.8) which is used to compute the meteorological variables that are needed to drive the

chemistry-transport model, and the MM5-CHIMERE model (http://www.lmd.polytechnique.fr/chimere/), developed by IPSL (Laboratoire de Météorologie Dynamique), which is used to predict the concentrations and deposition of several tropospheric species. MM5-CHIMERE has been primarily designed to produce daily forecasts of ozone, aerosols and other pollutants and to make long-term simulations for emission control scenarios. MM5-CHIMERE is executed over a range of spatial scales from a global and regional scale (domains of several thousands of kilometers) to an urban scale (100–200 km) with horizontal grid spacings of 1–100 km. Products are daily 72-h forecasts for O_3, NO_2, $PM_{2.5}$, PM_{10} and desert dust (http://prevair.org).

2.3.10 MM5/WRF-CMAQ (Spain, UK)

The CMAQ (United States Environmental Protection Agency, Community Multiscale Air Quality) model includes a suite of chemical as well as transport and dynamic schemes (Byun and Schere, 2006). It includes dynamical and chemical interactions between atmospheric pollutants on multiple scales in a modular framework. CMAQ has been designed for assessing the impact of multiple pollutants including tropospheric O_3 and other oxidants, speciated particulate matter, and acid-deposition species on time scales from an hour to years.

CMAQ is a widely used chemistry transport model which has been linked to a number of meteorological models including MM5, Eta and WRF (Sects. 3.2.8, 3.2.6, and 3.2.9, respectively). Although WRF has superseded MM5, the MM5-CMAQ is used for example to provide 72-h forecasts for hourly, daily maxima and daily average information related to O_3, NO_2, CO, SO_2, PM_{10}, $PM_{2.5}$ and NH_3 (http://verde.lma.fi.upm.es/cmaq_eu/).

Examples of CWF model forecasts can be found in Eder et al. (2006, 2009); they examine the performance of the model for forecasting 8-h ozone concentrations over the USA. CMAQ is also used operationally in the UK to predict footprints of industrial power plants for pollutants such as SO_2 and PM_{10} (e.g. Yu et al., 2008). CMAQ is supported and distributed by the Community Modelling and the Analysis System center (CMAS, http://www.cmascenter.org/).

2.3.11 MOCAGE (France, Spain, Romania)

The MOCAGE (Modèle de Chimie Atmosphérique à Grande Echelle, Model of Atmospheric Composition at Large Scales) three-dimensional multi-scale CTM has been designed at Météo-France for both research and operational applications. MOCAGE is applicable to CWF, tracking and back-tracking of accidental point-source releases, trans-boundary pollution assessment, assimilation of remote-sensing measurements of atmospheric composition, and studies on the impact of anthropogenic emissions of pollutants on climate change, with more than 40 references in the international peer-reviewed literature (e.g. Dufour et al., 2004; Bousserez et al., 2007).

MOCAGE has the flexibility to be run in different configurations with different parameterizations depending upon its application. The model considers the troposphere and stratosphere on the planetary scale and over limited-area subdomains at higher horizontal resolution. The model provides (by default) its own time-dependent chemical boundary conditions.

MOCAGE has been run daily since 2001. In 2004, Météo-France joined the partnership consortium and operational platform "Prév'Air" (http://www.prevair.org; Rouil et al., 2009) in charge of the pollution monitoring and forecasting for the French Ministry of Environment. Within this platform, 72-h forecasts are delivered daily to Prév'Air users, including ozone, precursors and aerosol over the globe (horizontal grid spacing of 2°), Europe (0.5°) and France (0.1°).

MOCAGE is also run daily in the context of GMES atmosphere projects (GEMS and now MACC, http://www.gmes-atmosphere.eu/services/raq), participating in the pre-operational ensemble forecasting system. The configuration used in this context has two domains and covers the globe at 2° horizontal grid spacing and Europe (15° W–35° E and 35° N–70° N) at 0.2° grid spacing. MOCAGE is also run by the Spanish and Romanian national meteorological services for their research and operations. MOCAGE is coupled to the computational fluid dynamics software PALM (http://www.cerfacs.fr/~palm) and can assimilate using variational methods (3DVAR, 3DFGAT or 4DVAR) profiles, columns or surface measurements of key atmospheric pollutants (e.g. El Amraoui et al., 2010).

2.3.12 NAME (UK)

NAME (Numerical Atmospheric dispersion Modelling Environment) is an off-line Lagrangian chemical transport model developed by the UK Met Office (Jones et al., 2007). It provides a flexible modelling environment able to predict dispersion over distances ranging from kilometres to the whole globe and for time periods from minutes upwards. This flexibility allows the model to be used in a variety of applications, including emergency response (e.g. Webster et al., 2007) and routine chemical weather forecasting.

NAME uses meteorology from the Met Office Unified Model (UM, Sect. 3.2.3) in either global or limited area configuration. Chemical modelling within NAME employs the scheme originally derived for the STOCHEM model (UK Meteorological Office Chemistry-transport Model, Collins et al., 1997). This scheme models gaseous and aqueous phase chemistry and has 40 advected and 18 non-advected tracers, 140 reactions and 23 photolytic reactions; 16 species are emitted. The dry deposition scheme used is based on a resistance network analogy for deposition velocity modelling. Particles can also be removed from the model atmosphere by fall out due to gravity, impaction with the surface, washout

by falling precipitation, and rainout, where the pollutant is absorbed directly into cloud droplets as they form.

Emissions are pre-processed using three datasets: (i) the UK National Atmospheric Emission Inventory at 1-km grid spacing; (ii) a 5-km grid-spacing inventory for shipping emissions around the UK; (iii) the EMEP 50-km grid spacing inventory outside the UK. The UK routine chemical weather forecast configuration of NAME employs a nested configuration, with an outer domain covering Western Europe and an inner domain covering the UK. The effective model grid for the inner domain corresponds to a grid spacing of around 8 km. The model provides forecasts out to 5 days and routine output parameters include atmospheric concentrations of ozone, CO, NO_2, SO_2, PM_{10} and secondary aerosol species.

2.3.13 OPANA (Spain and others)

OPANA is an Operational version of the ANA model (Atmospheric mesoscale Numerical pollution model for urban and regional Areas). OPANA is composed of several constituent models, including a nonhydrostatic mesoscale meteorological model (REMEST, based on MEMO and MM5, Sects. 3.2.7 and 3.2.8, respectively), a chemical model, emission model, and deposition model (San José et al., 2002). OPANA is designed to operate routinely to forecast air quality for 5–7 days. OPANA produces daily chemical weather forecasts in Leicester City Council (UK), Madrid Municipality (Spain) and Las Palmas de Gran Canaria (Spain). It is also used as an impact assessment system for industrial installations. Numerical products include daily 72-h forecasts for O_3, NO_2, CO, SO_2, and PM_{10} (http://artico.lma.fi.upm.es/).

2.3.14 RCG (Germany)

The urban-scale photochemical model CALGRID (California Grid Model; Yamartino et al., 1992) and the regional-scale model REM3 (Regional Eulerian Model; Stern, 1994) were used as the starting point for the urban/regional-scale model, REM-CALGRID (RCG, Stern, 2003). The RCG model has been designed to fulfill the requirements of the ambient air-quality framework directive 96/62/EC of the European Commission (Stern et al., 2008). RCG is run in offline mode. For long-term diagnostic applications, the meteorological driver is prepared by the analysis system TRAMPER (Tropospheric Realtime Applied Meteorological Procedures for Environmental Research, Reimer and Scherer, 1992).

For operational forecasting, the meteorology derives from the GME model (Global Model, Sect. 3.2.2). The model is part of a model system development including statistical and fuzzy models (Reimer and Dlabka, 2000) and Eulerian model RCG (Flemming and Reimer, 2000) to forecast especially surface ozone concentrations. The full system is documented in Reimer et al. (2000). After transformation of GME data to RCG coordinates, the boundary layer parameters are determined anew by the TRAMPER boundary-layer module.

The forecast system was developed for local abatement strategies for German authorities to apply the so-called German ozone law in the 1990s. With the introduction of EU directives, the diagnostic tests on emission scenarios became much more important than real-time forecasts. The RCG model has been operational for more than 10 yr, running every day at 12:00 UTC and producing a 72-h forecast over Central Europe (http://www.trumf.de/).

2.3.15 SILAM (Finland, Estonia, Russia, Lithuania and Spain)

The SILAM modelling system (Air-Quality and Emergency Modelling System) includes both Eulerian and Lagrangian dynamic kernels (e.g. Sofiev et al., 2006a, b; Saarikoski et al., 2007; Siljamo et al., 2008; Sofiev et al., 2009; Saarnio et al., 2010; http://silam.fmi.fi). The model applications range from global to meso-beta scale (grid spacing down to 1 km). The SILAM model is an open-access system, and the source code is publicly available on the web. The model is the official chemical weather forecasting tool on regional and larger scales in Finland and Lithuania. The model is also used for research purposes in Estonia, Russia, Lithuania and Spain.

The model has been used operationally in the EU-funded GEMS and MACC (http://www.gmes-atmosphere.eu) and EU-funded PROMOTE (http://www.gse-promote.org) and PASODOBLE projects. The predicted species include the concentrations of O_3, NO_x, SO_x, NH_x, VOCs, sea salt, anthropogenic $PM_{2.5}$ and PM_{10}, as well as pollution from wildland fires (e.g. Saarikoski et al., 2007; Sofiev et al., 2009, 2011b; Saarnio et al., 2010; and for selected biogenic aerosols, such as allergenic pollen (e.g. Sofiev et al., 2006b, 2011; Siljamo et al., 2008; Veriankaitë et al., 2010). The model is capable of four-dimensional variational data assimilation (Sofiev and Atlaskin, 2004). The model has recently been applied to evaluate the dispersion of primary $PM_{2.5}$ emissions across Europe and in more detail over Finland, and to assess the resulting adverse health impacts (Tainio et al., 2009, 2010; Karvosenoja et al., 2010).

The meteorological information is extracted most commonly from the Finnish Meteorological Institute (FMI) variant of the weather forecasting model HIRLAM (Sect. 3.2.4), which is used as a downscaling tool for ECMWF Integrated Forecast System forecasts (which are also used without modifications), and from the regional AROME (Applications of Research to Operations at Mesoscale, Sect. 3.2.5) model simulations for Southern Finland and the Baltic States. The products are 54- and 72-h forecasts for Finland, the Baltic States, and Europe (http://silam.fmi.fi). The structure of the SILAM modelling system has been schematically illustrated in Fig. 3.

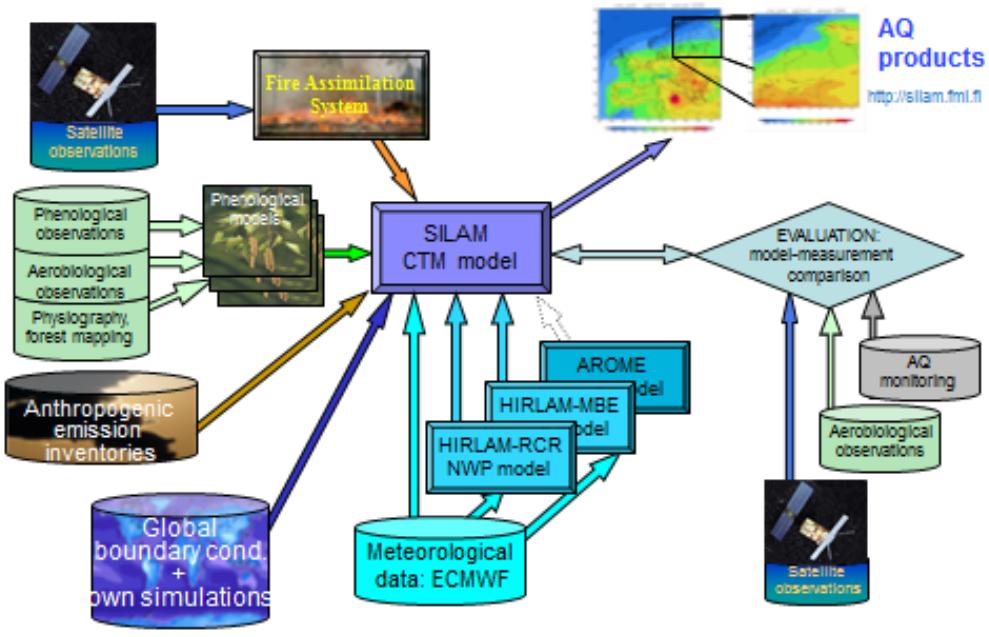

Fig. 3. The components, input and output data, and model evaluation of a regional chemical weather assessment and forecasting model SILAM.

2.3.16 SKIRON/Dust (Greece)

SKIRON/Dust is a modelling system that couples the National Oceanic and Atmospheric Administration (NOAA) Eta NWP model (Sect. 3.2.6) on-line with a dust transport model. As of this writing, SKIRON's NWP component runs at 5-km horizontal grid spacing, using the nonhydrostatic version with the Betts-Miller-Janjić convective parameterization scheme. Other modifications to the Eta model in SKIRON include a different radiative transfer scheme, differences in the soil properties in the surface parameterization, more soil and vegetation categories, and the incorporation of sloped surfaces in the surface energy balance.

The dust module of the system is based on the work of Nickovic et al. (2001) and Spyrou et al. (2010). The dust transport submodel includes eight size bins of dust particles (Marticorena and Bergametti, 1995; Zender et al., 2003; Pérez et al., 2006), the calculation of AOD, and the correction of radiative transfer through look-up tables (Kaufman et al., 2002). Dry and wet deposition schemes have been improved, and in-cloud scavenging has been included (Kumar et al., 1995; Seinfeld and Pandis, 1998; http://forecast.uoa.gr/dustindx.php).

2.3.17 THOR (Denmark)

THOR is an integrated air-pollution forecast and scenario management system (Brandt et. al., 2001b; http://thor.dmu.dk), consisting of an off-line coupled three-dimensional NWP model Eta (Sect. 3.2.6) and several air-pollution models (e.g. Danish Eulerian Hemispheric Model DEHM, Urban Background Model UBM, point source model OML, Operational Street Pollution Model OSPM, accidental release model Danish Rimpuff and Eulerian Accidental release Model DREAM). The model covers most of the Northern Hemisphere with a two-way coupled nest over Europe. The system is capable of 72-h forecasts of weather and air pollution from regional scale over an urban area down to individual street canyons in cities. DREAM can be used for any accidental release such as from power plants, industrial sites, and natural and human made fires (http://www2.dmu.dk/1_Viden/2_miljoe-tilstand/3_luft/4_spredningsmodeller/5_Thor/default_en.asp).

THOR can inform and warn the public in case of high air-pollution levels and for policy management (e.g. by emission-reduction or traffic scenarios) of many different chemical compounds. THOR is executed up to four times every day. The products are 72-h forecast and daily maximum of O_3, NO, NO_2, SO_2, and SO_4 for Denmark and Europe (http://www2.dmu.dk/1_Viden/2_Miljoe-tilstand/3_luft/4_udsigt/Default.asp).

2.3.18 WRF-Chem (Spain and others)

When the WRF (http://www.wrf-model.org/; Sect. 3.2.9) model is coupled with an atmospheric chemistry module to produce WRF-Chem (Grell et al., 2005) simulations of chemistry and aerosols from cloud scales to regional scales can be performed. WRF-Chem was developed by NCAR and NOAA with contributions from Pacific Northwest National Laboratory (PNNL), US Environmental Protection Agency

(EPA), and university scientists (http://www.acd.ucar.edu/wrf-chem/).

The version of WRF-Chem running in Spain is an online model; it solves the chemistry every 10 min and the meteorological time step is 5 min. The EMIssion MOdel (EMIMO) (Computer Science School – Technical University of Madrid, UPM) provides emission data for every grid cell, per hour and per primary pollutant, based on TNO European emissions. Products are daily 72-h O_3, NO_2, SO_2, CO, PM_{10}, $PM_{2.5}$ and NH_3 forecasts for Europe (http://verde.lma.fi.upm.es/wrfchem_eu/).

3 Numerical weather prediction models

Nearly all operational air-quality models have two components, a numerical weather prediction component and an air-pollution chemistry and physics component. The purpose of this section is twofold. First, we discuss the characteristics of NWP models that affect the ability of the coupled model to produce accurate forecasts of air quality (Sect. 3.1). Second, we provide a brief overview of the different NWP models in operational air-quality models in Europe (Sect. 3.2). Tribbia and Anthes (1987) provide a review of the scientific basis for numerical weather prediction, and Stensrud (2007) reviews physical parameterization schemes.

3.1 Selected characteristics of NWP models

In this section, the model architectures and physical processes in the NWP models are discussed.

3.1.1 Formulation of NWP models

NWP models can be divided into hydrostatic and nonhydrostatic models. Hydrostatic models assume that the accelerations of vertical velocity are small relative to that of gravity. Nonhydrostatic models, on the other hand, are capable of modelling vertical accelerations exceeding that of gravity, such as are found in deep, moist convection. Most models with horizontal grid spacings less than 10 km are run using nonhydrostatic models.

Another aspect is the vertical coordinate used in the formulation of the governing equations. Some models use pressure as their vertical coordinate, whereas others use a terrain-following σ coordinate, where $\sigma = (p - p_s) \div (p - p_t)$, p is pressure, p_s is surface pressure, and p_t is pressure at the top of the model (usually fixed at 100, 50, or 10 hPa). Others are hybrid systems that blend σ coordinates near the surface and pressure coordinates aloft, obtaining the benefits of both coordinate systems (simpler formulation of governing equations in pressure coordinates and better representation of near-surface flows along σ surfaces).

3.1.2 Cloud microphysical parameterizations

Accurate forecasting of the size distributions and number concentrations of cloud and precipitation water particles is important for deposition, photochemistry and aerosol-cloud-radiation interaction in CWF models. On the grid scale, cloud and precipitation processes are handled by cloud microphysical parameterizations. When grid-cell-sized regions of the model atmosphere become saturated, model clouds begin to form. Because these processes act on scales smaller than the grid-scale, cloud microphysical parameterizations handle the distributions and the conversions of condensed water substance between cloud water, rain water, cloud ice, snow, graupel, and hail. Schemes range in sophistication from (i) those that neglect ice processes to (ii) one-moment schemes (predicting only mixing ratios of each hydrometeor species) and to (iii) two-moment schemes (predicting number concentrations and mixing ratios). Because of the relative lack of knowledge about microphysical processes, higher-moment schemes and more sophisticated parameterizations do not necessarily lead to better predictions of clouds and precipitation. Straka (2009) presents a recent review of cloud microphysical parameterizations and principles.

3.1.3 Convective parameterization schemes

The choice of convective parameterization scheme in numerical weather prediction models is important for two reasons. As Baldwin et al. (2002) lament: "*All convective parameterizations contain arbitrary parameter settings and have characteristic behaviors that are sometimes inconsistent with reality*." First, the morphology and evolution of the convective systems that form in the model may depend on the convective scheme. For example, Bukovsky et al. (2006) showed that curved convective lines often formed in an operational model with the Betts-Miller-Janjić scheme, but did not initiate in the right place and time, whereas convective systems formed with the Kain-Fritsch scheme did not form bowing segments as frequently, but did initiate in the right place and time.

Second, how the convective parameterization changes the model atmosphere may not resemble what happens in reality. For example, Baldwin et al. (2002) showed that the Betts-Miller-Janjić scheme does not produce cold pools comparable to those observed in the wake of convective systems and may eliminate convective inhibition more quickly than in reality.

Models with horizontal grid spacings less than 5 km are often considered to be convection-permitting models, meaning that convective parameterizations can be omitted (at least partly for large-scale convection storms), allowing convective instabilities to be handled on the grid scale. In these models, the cloud microphysical parameterizations must do all the work of relieving instability. At least one study, however, has shown that improved handling of deep convection in

models in convection-permitting models can be achieved by running a convective parameterization (Yu and Lee, 2010). Another limitation of convective parameterization schemes is that only heat and moisture are redistributed. Momentum is generally not, partly because methods to handle such redistribution have not been developed.

A common assumption is that convective parameterization schemes exist to parameterize convective precipitation on the sub-grid scale. In fact, convective parameterization schemes exist to relieve the model of gravitational or buoyant instability in the vertical. Any precipitation created by the adjustment of the atmosphere back to stability because of the convective parameterization scheme (called convective precipitation or subgrid-scale precipitation) is merely a byproduct of the readjustment. Models usually have two schemes for releasing moist gravitational instability, one for deep convection such as thunderstorms, and one for shallow convection such as the stratocumulus clouds that cap the planetary boundary layer in the subtropics. Currently, of the models considered in this article, two convective schemes are implemented in the ECMWF IFS (Sect. 3.2.1) and Enviro-HIRLAM (Sect. 2.3.3).

There are two general classes of convective schemes. Because many convective parameterization schemes are developed from research on tropical convection, most of these schemes release the conditional instability almost as quickly as it is formed, maintaining convective neutrality. These are called statistical-equilibrium schemes, following the terminology in Emanuel (1994, Sects. 11.2 and 12.3) and Mapes (1997). Such schemes include the Kuo (1965, 1974), Arakawa-Schubert (1974), Betts-Miller (1986) and the Betts-Miller-Janjić (Janjić, 1994), and Tiedke (1989) schemes.

Convection in the mid and high latitudes, however, often does not behave in this manner. Instability may build up over hours or days, kept from being released by a lower-tropospheric stable layer or inversion called a cap or lid, measured by an energy barrier called the convective inhibition. Convection is released by some mechanism to lift unstable parcels past the layer of convective inhibition to release the instability. Such schemes are called activated or triggered schemes (Sects. 11.2 and 12.3 in Emanuel, 1994; Mapes, 1997). Such schemes include the Kain-Fritsch (Kain and Fritsch, 1990, 1993; Kain, 2004) scheme and its derivatives (e.g. Bechtold et al., 2001).

3.1.4 Boundary layer parameterization schemes for NWP models

Similar to convective parameterizations that redistribute heat and moisture when the atmosphere becomes unstable to moist processes, boundary-layer parameterizations serve a similar purpose for the near-surface layer of air. Some way is needed for models to distribute the heat, moisture, and momentum as the free atmosphere joins with the surface through the unresolved scales of turbulence present in the planetary boundary layer. The stability of the planetary boundary layer affects how this redistribution occurs, so some schemes work better for stably stratified situations (i.e. surface inversions), others work better for unstably stratified situations (i.e. convective boundary layers), and yet others work better for neutrally stratified situations (i.e. well-mixed boundary layers).

Despite their sophistication, the schemes used in NWP models have limitations, and these limitations can be critical for CWF applications. For example, CWF models may need greater vertical resolution within the boundary layer or improved surface sublayer parameterizations, especially for urban-scale air-pollution modelling where low-level emissions within the surface layer are occurring (from traffic, for example). Mixing height is a quantity needed for boundary-layer parameterizations, and it may be quite variable, especially over different land categories in urban areas or due to internal boundary layers, blending heights, etc. Furthermore, in some situations, the mixing height may even be poorly defined. Therefore, the boundary-layer parameterisations in NWP models used for CWF should be further improved, as discussed in the overview in Baklanov and Grisogono (2007) and Sokhi et al. (2010).

3.1.5 Initial and lateral boundary conditions

Initial conditions come from the observations collected worldwide and transmitted through the Global Telecommunications System. In addition, local sources of data such as Doppler radars, satellites, mesoscale observations, and buoys may also be included. The process by which observations are ingested into the model, interpolated onto the model grid, and balanced to produce a dynamically consistent set of initial conditions is called *data assimilation*. Some data assimilation systems are three-dimensional variational assimilation approaches (3DVAR), whereas others also include assimilating data in time (4DVAR). Some of the newest data-assimilation approaches involve ensemble Kalman filtering (EnKF), an approach that recognizes that the initial conditions are not (and can never be) perfectly known.

NWP models can cover a global domain or be limited-area models, with a regional domain. Limited-area models have to accommodate lateral boundary conditions from some larger-scale (usually global) model (the so-called noninteractive approach discussed by Staniforth, 1997). Because output from global models is infrequent (usually only stored every 3 or 6 h) and the limited-area models need input at their domain boundaries every model time step (usually tens of seconds), the data along the lateral boundaries is usually interpolated linearly in time. For situations where the flow may be changing or new features are moving into the limited-area domain through the boundaries, large errors may be introduced (Nutter et al., 2004; Elmore et al., 2006b).

To accommodate for boundary issues, noise, model spin-up, and other specious features entering the domain, an adjustment region is required upstream of the relevant

meteorological region. Staniforth (1997, p. 19) offers a simple practical test that limited-area models should meet: "*the solution obtained over a limited area should well match that of an equivalent-resolution model integrated over a much-larger domain.*" Alternatively, an interactive approach could be employed either through two-way nesting or variable resolution (Staniforth, 1997).

3.2 NWP models used in European CWF modelling systems

In this section, we discuss the different NWP models that commonly are used to provide meteorological data to the operational chemical weather models. The characteristics of each model are summarized in Table 2. Rather than in alphabetical order, the models are discussed in two groups: hydrostatic models then nonhydrostatic models.

3.2.1 ECMWF IFS

Widely regarded as the most accurate NWP model in the world, the ECMWF was developed from a European COST action to provide global medium-range weather forecasts (Woods, 2006). At the time of this writing (August 2011), the nonhydrostatic ECMWF model known as the Integrated Forecast System (IFS) is T1279, an equivalent grid spacing of about 16 km, with 91 vertical hybrid-coordinate levels and a model top at 0.01 hPa. The model is run twice a day at 00:00 and 12:00 UTC. The nonhydrostatic dynamical core comes from the ALADIN model (Sect. 3.2.5). The convective scheme is a modified Tiedke (1989) scheme (Nordeng, 1994), testing for shallow, deep, and midlevel convection (e.g. above a frontal zone or inversion). The surface scheme is Tiled ECMWF Scheme for Surface Exchanges over Land (TESSEL) and has been revised to include surface hydrology and the choice of a global soil texture (Balsamo et al., 2009). Access to the model and its output is proprietary for 18 European member states and 10 states with co-operation agreements.

3.2.2 GME

GME (global model) from German Weather Service (DWD) is a hydrostatic model and is designed for the routine forecast of complex weather development on synoptic scales (Majewski et al., 2002). The vertical domain extends up to the stratosphere. For regional weather forecasts, the nonhydrostatic limited-area models COSMO-EU and COSMO-De (Consortium for Small-Scale Modelling) are used with boundary conditions from GME (www.dwd.de). The global model GME is defined on an icosahedral grid with about 60-km horizontal grid spacing. Within the postprocessing, all fields are transformed to a latitude-longitude geographical grid.

3.2.3 Unified model

The Unified Model (UM; Cullen, 1993) was developed by the UK Met Office and introduced in 1990. The Unified Model can serve as a hydrostatic global model or a nonhydrostatic limited-area model (Davies et al., 2005). The model vertical coordinate is height, and the convective parameterization is based on Gregory and Rowntree (1990). The boundary-layer scheme is described by Brown et al. (2008). The Unified Model is a proprietary model and has limited access to those outside the UK Met Office.

3.2.4 HIRLAM

The High-Resolution Limited Area Model (HIRLAM; Undén et al., 2002; http://hirlam.org) derives from a consortium of European meteorological institutes (Denmark, Estonia, Finland, Iceland, Ireland, Norway, The Netherlands, Spain, and Sweden, with France as a research partner). HIRLAM is run at a variety of grid spacings from 50 to 1.5 km, depending on country, with the reference version being run at FMI. HIRLAM is a hydrostatic model, although a nonhydrostatic version also exists. Three convective parameterization schemes are available: STRACO (Soft TRAnsition COnvection), which is a modified Kuo scheme that aims to produce a smooth transition between convective clouds and large-scale condensation, Rasch and Kristjánsson (1998), and Kain and Fritsch (1990, 1993). HIRLAM is available to member states, but access can be granted for other users by a special agreement.

3.2.5 ALADIN, ARPEGE, and AROME

ALADIN (Aire Limitée Adaptation Dynamique INitialisation) is a limited-area version of the French global model ARPEGE (Action de Recherche Petite Echelle Grande Echelle, which was the basis for the ECMWF IFS), growing out of a French-led consortium. ALADIN uses a modified version of the Bougeault (1985) convective scheme and a terrain-following-pressure hybrid vertical coordinate. The ALADIN and HIRLAM consortia joined together starting in 2004 to form the HARMONIE consortium (Hirlam Aladin Research on Meso-scale Operational NWP in Euromed) in which a new model has been developed (AROME; Seity et al., 2010).

3.2.6 Eta

The Eta model was the operational limited-area hydrostatic model from June 1993 to June 2006 in the United States. The model uses a unique step-coordinate vertical coordinate called the eta (hence the name of the model), a modified version of the σ coordinate (Mesinger et al., 1988; Janjić, 1990, 1994). The Eta model uses a version of the Betts-Miller convective scheme (Betts and Miller, 1986; Janjić, 1994; Baldwin et al., 2002). Research versions of the Eta included

a sigma-coordinate version, a nonhydrostatic version, and a version with the Kain-Fritsch convective parameterization scheme (Baldwin et al., 2002; Kain et al., 2003). One problem with the Eta model is that it fails to reproduce strong surface winds associated with downslope windstorms, partly as a result of the way that the mountain waves are handled in the eta coordinate system (Gallus, 2000; Gallus and Klemp, 2000).

3.2.7 MEMO

MEMO (MEsoscale MOdel) is a nonhydrostatic mesoscale model for simulating wind flow. MEMO was developed by the Aristotle University of Thessaloniki and the University of Karlsruhe. MEMO has been used to study the wind fields around urban areas (e.g. Kunz and Moussiopoulos, 1995). The model uses a terrain-following coordinate and has two physical parameterizations of importance – radiative transfer (Moussiopoulos, 1987) and K-theory for the planetary boundary layer – but neglects moist processes by assuming the atmosphere is unsaturated. MEMO is coupled with the photochemical chemical transport model MARS (Model for the Atmospheric Dispersion of Reactive Species) to produce the European Zooming Model (EZM; Moussiopoulos, 1995). Because MEMO does not contain moist atmospheric processes, its applicability when clouds and precipitation are present is limited.

3.2.8 MM5

The fifth-generation Pennsylvania State University/National Center for Atmospheric Research Mesoscale Model (MM5) is a nonhydrostatic limited-area model that has been one of the most popular open-source mesoscale models in the world (Dudhia, 1993; Grell et al., 1994; http://www.mmm.ucar.edu/mm5). MM5 is a terrain-following σ-coordinate model with a large degree of flexibility in choosing domains, nestings, grid spacings, and model physics. For example, as of the writing of this article, users of the MM5 can choose from six convective parameterizations, seven resolvable-scale cloud microphysics parameterizations, six planetary boundary-layer parameterizations, seven surface parameterizations, and four atmospheric radiation schemes, although many of these schemes are outdated, overly simple, or inappropriate for some weather situations. As such, care is needed when configuring the model to ensure optimal performance.

3.2.9 WRF

The successor to the MM5 is the open-source Weather Research and Forecasting model (WRF; Skamarock and Klemp, 2008; Wang et al., 2009). The goal of WRF is to produce a common architecture for both research and operations to build upon (http://wrf-model.org). WRF has two nonhydrostatic dynamic cores, the ARW (Advanced Research WRF), developed primarily by the National Center for Atmospheric Research, and the NMM (Nonhydrostatic Mesoscale Model; Janjić et al., 2001; Janjić, 2003), developed primarily by the NOAA/National Centers for Environmental Prediction. The WRF-ARW uses a σ vertical coordinate in either a limited-area or global domain, whereas the WRF-NMM uses a hybrid σ-pressure vertical coordinate. One of the recent additions to WRF is the positive-definite advection scheme, which improves the conservation of advected quantities and prevents negative quantities such as mixing ratio and chemical concentrations (Skamarock, 2006; Skamarock and Weisman, 2009).

3.2.10 RAMS

The Regional Atmospheric Modeling System (RAMS; Pielke et al., 1992; Cotton et al., 2003) is a nonhydrostatic limited-area model developed by Colorado State University. Like MM5, RAMS is a terrain-following σ-coordinate model with the flexibility to choose the domains, grid spacings, and model physics packages.

4 Chemical transport models: architectures and physical processes

The chemical transport models (CTM) simulate processes controlling the distribution of chemical species in space and time. To calculate atmospheric concentrations of pollutant species, the modelling framework needs to incorporate several key processes: advection (Sect. 4.1), turbulent diffusion (Sect. 4.2), chemical transformation (Sect. 4.3), aerosol processes and microphysics (Sect. 4.4), deposition of pollutants (Sect. 4.5), temporal allocation and distribution of anthropogenic and natural emissions (the latter in Sect. 4.6), and horizontal and vertical grid spacing (Sect. 4.7). Although there are a large number of three-dimensional CWF models available, most of these are based on similar frameworks for linking these interactions, and they all solve the continuity equations for mass conservation of the pollutants in the atmosphere. Anthropogenic emission inventories have not been addressed in detail in this study, although some challenges in their development are discussed in Sect. 7.1.

Transport of pollutant species involves both advection and diffusion. Advection refers to the movement of pollutant species by the mean wind fields, whereas diffusion involves sub-grid-scale turbulent mixing of pollutants. By definition, advection transports the pollutants without a significant change in the concentration in the considered volume, whereas diffusion involves dilution and hence leads to lowering of pollutant concentrations. In an Eulerian frame of reference, the computational domain of a CWF model consists of a matrix of contiguous grid cells forming a finite three-dimensional volume (also called a box). As this box

is a subset of the entire atmospheric globe, lateral boundary conditions define the advection into the modelling grid.

Horizontal advection is typically considered dominant, and no exchange at the top boundary of the domain is assumed. However, a realistic representation of stratospheric intrusions into the troposphere is possible only by adjusting the boundary conditions at the top. The model also has to include at least the lower stratosphere to allow for these usually small-scale features. Consequently, stratospheric intrusions are not properly addressed by the currently available operational models.

One of the key problems in atmospheric composition modelling is knowing the accuracy and reliability of the numerical schemes applied. A less appreciated, but also an important, issue is to ensure the compatibility between the schemes applied in different modules of the modelling system. Usually, the model construction employs operator splitting (e.g. Seinfield and Pandis, 2006), thus distinguishing the advection scheme, diffusion algorithm, chemical transformation module, dry, and wet deposition, data assimilation control module, and a set of supplementary modules including meteorological pre-processor. Of these, advection and diffusion are usually closely linked. The order of evaluating the terms can influence the results. Therefore, the half-step mirroring is used sometimes. In other words, all terms are evaluated twice during each model time step – first in direct and then in reverse order. This approach may also increase the order of approximation (Marchuk, 1986).

The main physical and chemical components of the selected 18 models have been summarized in Table 3.

4.1 Advection

Existing advection schemes can be categorized by one of four approaches: finite-difference, flux, semi-Lagrangian, and spectral. The basic principles of these schemes were formulated several decades ago and, with certain modifications, are still applied.

The diversity of advection routines developed during the last 50 yr and still under construction is explained by a long list of requirements to such schemes. The most important ones are positive definiteness to the scheme, minimal numerical viscosity, limited non-monotonicity, sufficiently high stability, absence of phase error, local and global mass conservation, and sufficiently high numerical efficiency. Unfortunately, meeting all requirements simultaneously is not possible. For example, more numerical viscosity smoothes the result, thus improving monotonicity.

The most important criteria when selecting a scheme seems to be the positive definiteness of the algorithm (i.e. a guarantee that mass will remain positive after the advection step) and monotonicity (i.e. minimizing high-frequency fluctuations of the field). These two criteria can be optimised to some extent at a cost of substantial numerical viscosity, which is a common problem for most of the Eulerian advection schemes. Only the scheme of Galperin (1999, 2000) has exactly zero numerical viscosity, but at a somewhat increased non-monotonicity of instant concentration fields and additional memory requirements.

Other criteria, sometimes mentioned but rarely given the highest priority, are minimization of phase error (i.e. correct representation of the transport velocity), the scheme transportivity (i.e. shift of the centre of mass of a puff from the analytical solution), and additivity (i.e. correct treatment of superimposed puffs). Two other features are also important: the conservation of mass and costs of the computations. The mass-conservation problem is usually considered as the highest priority in the chemistry-transport models, so that schemes that significantly violate this requirement are excluded from consideration. Finally, the efficiencies of the advection schemes (both in terms of the computational time and memory) are potentially important, but rarely considered more important than positive definiteness or mass conservation.

Interestingly, these two features are not the most important aspects to be considered for NWP models. Although the conservation of mass is desirable in NWP models, this criterion is usually compromised, if an algorithm that is not exactly conservative has a better performance or monotonicity.

The finite-difference schemes involve direct discretization of the dispersion equation and involve various types of interpolation functions to evaluate derivatives of the concentration fields, as reviewed by Richtmyer (1962), Leith (1965), and Roach (1976). Examples of specific developments are van Leer (1974, 1977) and Russel and Lerner (1981). These once-popular schemes usually suffer from substantial numerical viscosity and limited stability, which sets very stringent limitations to the Courant number (the ratio of the maximum distance passed during the model time step to the model grid cell size). Consequently, interest has largely shifted towards flux and semi-Lagrangian schemes for practical applications.

Flux-type schemes are based on an evaluation of the admixture fluxes at the borders of the grid cells using some interpolation procedure for determining the concentrations and wind speed (e.g. Odman, 1998). Probably the most widely used flux-type scheme is Bott (1989, 1992, 1993) and its derivatives (e.g. Syrakov, 1996; Syrakov and Galperin, 1997, 2000) involving different approximation functions (i.e. Bessel functions instead of Lagrangian polynomials). Although these schemes suffer from both stability and viscosity problems, they are superior to finite-difference approaches. Flux-type schemes also require special efforts to maintain mass conservation.

Semi-Lagrangian schemes (e.g. Crowley, 1967, 1968; Egan and Mahoney, 1972; Pedersen and Prahm, 1974; Smolarkiewicz, 1982; Prather, 1986; Williamson and Rasch, 1989; Staniforth and Côté, 1991, and references therein; Galperin, 1999, 2000; Sofiev, 2000b) represent the concentrations as a set of masses distributed according to certain rules inside the grid cell and advect like Lagrangian particles,

although some properties are conserved. A sub-class of these schemes include purely Lagrangian schemes (Eliassen, 1978; Eerola, 1990; Stohl et al., 2005) where the masses are essentially independent and transported individually with the follow-up reprojection to the computational grid. These schemes have better numerical viscosity, which can be made exactly zero (Galperin, 2000), and stability (i.e. their working range of Courant number is probably the widest out of all types of the advection schemes). However, many semi-Lagrangian schemes exhibit large non-monotonicity and other distortions of the transported field, which require substantial efforts and computational resources to be kept under control.

By comparison, Lagrangian schemes are rarely used for chemistry composition computations, mainly due to the unbearable overhead of meeting the requirements posed by nonlinear chemical transformation mechanisms. Nevertheless, such schemes are theoretically possible. Such schemes naturally resolve the numerical diffusion problem, which plagues the performance of almost all Eulerian schemes. Although the diffusion problem is seemingly inevitable in Eulerian schemes, Lagrangian advection schemes do not suffer this problem because they lack explicit discretization of horizontal movement, which is performed in continuous space rather than in predefined grid meshes. As a result, numerical viscosity of purely Lagrangian schemes is always zero. Such a result comes at a price of 100 % non-monotonicity of the concentration fields, which originates from limited spatial representativeness of a single Lagrangian particle.

Spectral models (e.g. Kreiss and Oliger, 1972; Prahm and Christensen, 1977; Zlatev and Berkowicz, 1988) use Fourier transformation to convert the differential equations into algebraic ones, which are then solved analytically. Such schemes are more commonly used in NWP models than chemical transport models.

One new line of development, often based on semi-Lagrangian schemes, is adaptive-grid advection algorithms (e.g. Staniforth and Cote, 1991 and references therein; Lagzi et al., 2004; Jablonowski, 2004 and references therein; Jablonowski et al., 2006). These schemes are geared at solving the problems with sharp gradients in the computed variables and with a wide range of spatial and temporal scales of input forcing. The advantage of more accurate computations in the sub-domains that require high resolution outweighs the extra errors introduced by repeated reprojection of the main fields, as well as the extra computational time needed for the grid transformation.

Within the 18 models considered in this review, only a few use the same type of advection scheme. There is, however, a general lack of detail regarding the descriptions of the advection mechanisms in the publications for each of the models, which suggests that the schemes are implemented with minor, if any, deviations from the original source. Examples of the schemes used by the selected models are: Bott (1989) (CAMx, Enviro-HIRLAM, MATCH), Piecewise Parabolic Method (PPM) of Colela and Woodward (1984) (CAMx, CMAQ and CHIMERE), Walcek (2000) (RCG, LOTOS-EUROS), Yamartino (1993) and Wicker and Skamarock (2002) (CMAQ, WRF-Chem and FARM), Williamson and Rasch (1989) (semi-Lagrangian scheme of MOCAGE), Eerola (1990) (Lagrangian SILAM kernel), Galperin (1999, 2000) (Eulerian SILAM kernel), Janjić (1997) (SKIRON/Dust), Zlatev (1995) (THOR), and Cullen (1993) (NAME).

Some models have more than one advection algorithm. In particular, CAMx allows the user to choose between Bott and PPM, as well as an optional Plume-in-Grid formulation for representing the large point sources in the lower-resolution grid. Enviro-HIRLAM has options for semi-Lagrangian schemes, in particular the CISL (Kaas, 2008). SILAM has two dynamic kernels: Lagrangian and Eulerian; either can be selected for a particular run via a switch in the model control file.

4.2 Horizontal and vertical turbulent diffusion

Turbulent mixing representation is substantially different for meteorological and chemical transport parts of the operational chemical weather models. For meteorological modules, the role of turbulence is more important, as it is the turbulent friction that connects the model to the surface, ensures exchange of momentum in the vertical and, in the end, leads to formation of the planetary boundary layer. In the chemical transport models, the role of turbulence is limited to an additional term of the tracer transport, which is usually important only along the vertical axis.

The basics of the so-called turbulent closure for the atmospheric dynamics models was laid down by Boussinesq (1877), who introduced the term of eddy viscosity as a means of description of the momentum flux due to presence sub-grid-scale (unresolved) turbulent eddies and grid-scale (resolved) gradient of wind speed. The approach was further generalised to the transport of any scalar quantity by introducing the eddy diffusivity connecting the grid-scale scalar flux and its gradient. Smagorinski (1963) suggested a useful formula for eddy viscosity in numerical models, which is based on local derivatives of the wind speed and the model resolution. It is still used by many atmospheric dynamics models.

Some of the complexities of implementing diffusion are the limited applicability of the first-order Boussinesq closure in both strongly stable and convective stratifications, problems in adequate description of the turbulent length scale in the free troposphere (i.e. where the distance to the surface is no longer an adequate scale), and a "competition" with the numerical viscosity of the advection schemes. Strong stratification leads to the appearance of significant anisotropy of turbulence and correlation between the orthogonal directions. Some extreme cases, such as deep convection, can be

taken out of the diffusion module and parameterised separately (Sect. 3.1.3).

The horizontal diffusion is based on the Smagorinsky approach (1963) within ALADIN-CAMx, SKIRON/Dust, FARM, CAMx-AMWFG and MM5-CAMx. MM5-CHIMERE and MM5-CAMx use the Medium-Range Forecast Planetary Boundary Layer (MRF PBL) scheme. Horizontal diffusion in the CMAQ model is based on a grid-size-dependent algorithm that combines Smagorinsky's approach with a term to minimize numerical diffusion (Byun and Schere, 2006). MM5 uses several PBL schemes: bulk, high-resolution Blackadar, Burk-Thompson, Eta, Gayno-Seaman and Pleim-Chang. SILAM involves two approaches, depending on the kernel. The solution with the Lagrangian kernel uses prescribed horizontal diffusion via random particle relocation at each model time step (Eerola, 1990). In contrast, the Eulerian one includes an embedded algorithm that reflects the main features of the K-closure model; the horizontal eddy diffusivity is dependent on the wind speed.

For the vertical diffusion, the K-diffusion scheme is widely used, but otherwise, there are not many similarities between different models. In ALADIN-CAMx and MOCAGE, the vertical diffusion is calculated according to the Louis (1979) approach, which uses the Richardson number and the mixing length. Two convective boundary-layer schemes are implemented in OPANA: Blackadar and the Asymmetric Convective Model (ACM, Pleim and Chang, 1992). OPANA also includes local diffusion, vertically continuous integration, smooth transition from stable to convective with a faster matrix solver, and an updated eddy diffusion scheme.

RCG's vertical turbulent mixing formulation uses K-diffusion in combination with mixing height, which is treated as one layer above a 50-m surface layer. Its stable and convective boundary-layer diffusion coefficients are based on PBL scaling regimes. Therefore, vertical mixing is dominated by the time-dependent evolution of the mixed layer.

FARM also uses K-diffusion; eddy viscosities can be produced by the meteorological driver or by the preprocessor SURFPRO (SURrface atmosphere interFace PROcessor), which can choose among different parameterisations, based on PBL scaling. In MM5-CHIMERE, the vertical turbulent mixing takes place only in the boundary layer. The formulation uses K-diffusion, without the counter-gradient term. The vertical diffusion is mainly modelled with the ACM2 (Pleim, 2007a, b) in MM5-CMAQ; WRF-Chem uses the PBL parameterization by the Yonsei University (YSU).

The Eulerian kernel in SILAM is modelled according to the K-closure, with the approach of Genikhovich et al. (2004) used for the evaluation of K_z. The Lagrangian kernel within SILAM assumes a well-mixed PBL and fixed random-walk parameters in the free troposphere. Exchange between the PBL and the troposphere in the Lagrangian version takes place due to variation of the PBL height. In MATCH, the turbulence is parameterized using three primary parameters: the surface friction velocity, the surface sensible heat flux and the boundary-layer height.

The main limitation of the Lagrangian system of SILAM is the assumption of a well-mixed PBL. For the Eulerian SILAM variant, the K-closure is used for diffusion in the vertical direction and also for horizontal diffusion. The eddy diffusivity of the vertical profile is evaluated at every time step by Sofiev et al. (2010). The limitations on the large scale originate partly from the simplified free-troposphere diffusion. The Lagrangian kernel assumes fixed mixing coefficient, whereas the Eulerian one assumes 10 % of the PBL maximum K_z value. In LOTOS-EUROS, the mixing layer is treated as one layer, and the ground level output is generated by assuming a vertical profile near the ground based on the deposition velocities.

4.3 Chemistry

Presently the main air-pollution issues in Europe are the human health impacts of exposure to particulate matter and ozone, and to a lesser extent nitrogen dioxide, sulphur dioxide, carbon monoxide, lead and benzene (EEA, 2007). Ozone is formed in the atmosphere in photochemical reaction cycles, which brings the ozone precursors (i.e. NO_x, VOCs) and their gas-phase atmospheric chemistry to the focus of CWF. All state-of-the-art chemical modules in most of the modelling systems in this review include these reaction cycles.

The choice of a chemical scheme for a CWF model is always a compromise between its complexity, the requirements and restrictions of the modelling system, how it is applied, and the available computational resources. Basic gas-phase inorganic chemistry is usually included in all models, and the schemes are often quite similar because inorganic atmospheric chemistry is well established.

A photochemical oxidation mechanism of VOCs is necessary in any CWF model aiming to predict the ozone concentrations – here the models may differ considerably, with different levels of detail and different parameterisations. In addition to the anthropogenic VOCs, the oxidation of biogenic VOCs should be included, especially if the model domain covers regions with dense forests or agricultural lands. If the modelling system is to be used for acid deposition or acidification/eutrophication studies, representation of aqueous-phase sulphur chemistry is required, and, to predict the concentrations of atmospheric particulate matter, a representation of inorganic gas-particle partitioning is needed. Several approaches have been developed, which typically involve the simplification of more comprehensive chemical schemes to include only the key chemical constituents and processes. In the following, concise descriptions of some viable approaches are given.

The most commonly used chemical sub-model types among the chemical weather prediction models discussed in this review are (in alphabetical order): CBM-IV, RADM and

Table 4. Selected main characteristics of the chemical submodels that are included in the CWF models. NC = not considered.

Chemical sub-model	Dispersion models	Chemical species	Photolysis rates	Aqueous-phase	References
Acid-Basic	SILAM	29 species (18 advected)	12 reactions	Oxidation of SO_2 to sulphates, parallel to gas-phase reaction	Galperin and Sofiev (1998); Sofiev (2000)
CBM-IV (CB-IV)	CAMx CMAQ LOTOS-EUROS OPANA RCG	33 compounds 81 reactions	12 reactions. For most of the species the clear sky photolysis rates are calculated according to the Roeths flux algorithm	NC	Gery et al. (1989)
ISORROPIA	CAMx CHIMERE CMAQ LOTOS-EUROS RCG	22 species 17 equilibrium reactions	NC	NC	Nenes et al. (1998a,b) New version: http://nenes.eas.gatech.edu/ISORROPIA/ Fountoukis and Nenes (2007)
MELCHIOR	CHIMERE	Extended mechanism: 80 compounds 320 reactions (26 inorganic) Reduced mechanism: 44 compounds 133 reactions (26 inorganic)	22 photolysis reactions Photolysis rates calculated under clear sky conditions as a function of height and attenuated by cloudiness	Aqueous oxidation of SO_2 by O_3, H_2O_2, NO_2 and O_2 catalyzed by metal ions	http://www.lmd.polytechnique.fr/chimere Schmidt et al. (2001)
NWP-Chem	Enviro-HIRLAM	17 (28) advected species 27 (32) gas-phase reactions	4 photolysis reactions	Simplified liquid-phase equilibrium mechanism NWP-Chem-Liquid with 13 reactions	Korsholm et al. (2008) Korsholm et al. (2009)
RACM	Enviro-HIRLAM EURAD MOCAGE	77 compounds 214 reactions	23 photolysis reactions, procedure the same as in RADM2	NC	Stockwell et al. (1997)
RADM2 (RADM)	CAMx CHEM CMAQ Enviro-HIRLAM EURAD OPANA WRF-Chem	63 compounds 156 reactions (38 inorganic)	21 photolysis reactions The photolysis module uses a radiative transfer model. This module calculates photolysis frequencies that considers changes in the radiation with height and changes in air composition such as O_3; aerosols and water vapor		Stockwell et al. (1990)
SAPRC-99	Aladin-CAMx CMAQ FARM OPANA	80 compounds 214 reactions (48 inorganic) 16 radicals	24 photolysis reactions Rate constants must be calculated from their corresponding absorption cross sections and quantum yields given the spectrum and intensity of the sunlight or other light source in the simulation	NC	SAPRC-99: http://www.engr.ucr.edu/~carter/SAPRC99.htm SAPRC-07: http://www.engr.ucr.edu/~carter/SAPRC
UNI-OZONE	EMEP MATCH(EMEP-MSC-W)	71 compounds 123 reactions (22 inorganic) 24 radicals (Ozone concentrations from two-dimensional global model scaled by observed total ozone colums)	22 photolysis reactions J-values calculated over clear sky conditions and for two predefined clouds	Aqueous oxidation of SO_2 by O_3, H_2O_2 and O_2 catalyzed by metal ions	http://www.emep.int/OpenSource/index.html Simpson et al. (2003)

RACM, SAPRC-99, and UNI-OZONE. In addition, three other chemical schemes, each implemented in only one of the discussed CWF systems, are considered: MELCHIOR, NWP-Chem, and SILAM acid basic. Some characteristics of these chemical sub-models are compared in Table 4.

Carter's (1996) one-product isoprene oxidation scheme is adopted for biogenic compounds in several models, and the ISORROPIA thermodynamic equilibrium scheme (http://nenes.eas.gatech.edu/ISORROPIA/, Nenes et al., 1998a, b) is used to determine the physical state and composition of inorganic aerosols in many modelling systems. ISORROPIA does not consider aerosol size distributions or aerosol microphysical processes, which is why it is included in this section with other purely chemical schemes, instead of in Sect. 4.4.

The implementation of the chemical mechanisms in the chemical weather prediction systems often involves adaptations, updates or other modifications of the original scheme. These changes are not always well documented or

Table 5. Comparison studies of the various chemical sub-models.

References	CBM-IV (CB-IV)	ISORROPIA	MELCHIOR	RADM2 (RADM)	SAPRC-99	UNI-OZONE (EMEP)	Other not discussed chemical submodules
Anderson-Sköld and Simpson (1999)						X	X
Ansari and Pandis (1999a, b)		X					X
Cuvelier et al. (2007)	X	X	X		X	X	X
Dodge (2000)	X			X	X		X
Faraji et al. (2008)	X				X		
Gross and Stockwell (2003)				X		X	X
Jimenez et al. (2003)	X			X	X	X	X
Kuhn et al. (1998)	X			X		X	X
Luecken et al. (2008)	X				X		X
Sarwar et al. (2008)	X						X
Tilmes et al. (2002)	X			X		(X)	X
van Loon et al. (2007)	X	X	X			X	X
Vautard et al. (2007)	X	X	X		X	X	X
Yu et al. (2005)		X					X

N.B. In some studies EMEP refers to older versions of the EMEP model chemistry.

transparent. Therefore, only the general features of the original chemical schemes are discussed below. In the following, the models using the different chemical modules are listed in brackets in the titles. More information about the details of the implementation of any chemical submodule in a particular CWF system can be found at the web sites of the prediction systems. A list of references on the comparisons of various chemical submodules is presented in Table 5.

4.3.1 Acid basic (SILAM)

The scheme is a further development of the DMAT model (dispersion model for atmospheric transport) algorithm (Pressman et al., 1991; Galperin and Sofiev, 1998; Sofiev, 2000) and it treats the production processes of secondary inorganic aerosols, such as sulphates, nitrates and ammonia. It includes 29 species, 12 photochemical, 27 inorganic and 12 methane and ethane reactions. Most of reactions take place in the gas phase and constitute the oxidation of SO_2, NO_x, and NH_3.

The ozone cycle is considered via the photostationary equilibrium shifted in the presence of organic species. This approach does not lead to accurate ozone estimates, but is sufficient for partitioning NO_x into NO and NO_2. Aqueous-phase and heterogeneous reactions are responsible for within-droplet SO_2 oxidation, N_2O_5 hydrolysis and three-component equilibrium between ammonium, ammonium nitrate, and nitric acid, the description of which generally follows Finlayson-Pitts and Pitts (1998).

The previous version of the scheme has been evaluated within the scope of the EMEP programme (Sofiev et al., 1994), and a multi-annual evaluation was made by Sofiev (2000). The current version of the scheme is used in the MACC project with daily operational evaluation (http://www.gmes-atmosphere.eu). A comparison with NO_2 total column observed by Ozone Monitoring Instrument (OMI) onboard of Aura NASA (National Aeronautics and Space Administration) spacecraft has been performed by Huijnen et al. (2010).

4.3.2 CBM-IV (CAMx, CMAQ, ENVIRO-HIRLAM, LOTOS-EUROS, OPANA, RCG, SILAM)

The Carbon bond mechanism IV (CBM-IV, also called CB-IV; Gery et al., 1989) is a lumped-structure condensed mechanism. The carbon bond approach is used to lump organic species. The code treats the reactions of four different types of species: inorganic species, explicit organic species, organic species represented by carbon surrogates and organic species that are represented by molecular surrogates. Inorganic chemistry is represented explicitly with no lumping. Organics represented explicitly are formaldehyde, ethene and isoprene. Carbon bond surrogates describe the chemistry of different types of carbon bonds commonly found as parts of lager molecules. CBM-IV is widely used in research and regulatory chemical weather models, such as Models-3/CMAQ (Byun and Ching, 1999). Recently, an updated version of the Carbon Bond mechanism (CBM05) has become available, and has been implemented in the most current versions of the CMAQ model (Yarwood et al., 2005; Sarwar et al., 2008; Luecken et al., 2008). Among other changes, this version of the mechanism contains 156 reactions involving 52 chemical species, updated rate constants, an extended inorganic reaction set for urban to remote tropospheric conditions, and NO_x recycling reactions to represent the fate of NO_x over multiple days.

4.3.3 ISORROPIA (CAMx, CHIMERE, CMAQ, ENVIRO-HIRLAM, LOTOS-EUROS, RCG, FARM)

ISORROPIA ("equilibrium" in Greek) is a thermodynamic equilibrium aerosol module designed for the calculation of equilibrium concentrations of semi-volatile inorganic species (Nenes et al., 1998a, b; Fountoukis and Nenes, 2007). The aerosol system consists of sulphate, nitrate, ammonium, sodium, chloride and water, partitioned between gas, liquid and solid phases. Aerosol particles are assumed to be internally mixed (i.e. all particles of the same size have the same composition), and the model also determines the water content of the particles. In ISORROPIA, four distinct chemical species are possible in the gas phase, twelve in the liquid phase and nine in the solid phase. The number of species and equilibrium reactions solved in the calculation is determined by the relative abundance of each aerosol precursor (NH_3, Na, HNO_3, HCl, H_2SO_4) and the ambient relative humidity and temperature.

ISORROPIA has been optimized for speed and robustness for application in urban, regional and global chemical weather models. The performance of ISORROPIA has been evaluated against several in-situ datasets (e.g. Nowak et al., 2006) and compared to other thermodynamic equilibrium schemes (e.g. Nenes et al., 1998b; Ansari and Pandis, 1999a, b; Yu et al., 2005). A new version of the ISORROPIA module that includes the treatment of the crustal species (Ca, K, Mg) is called ISORROPIA II (not used at the moment in any of the discussed CWF models), and it is available online at http://nenes.eas.gatech.edu/ISORROPIA/.

4.3.4 MELCHIOR (CHIMERE)

The MELCHIOR (Modele Lagrangien de Chimie de l'Ozone a l'échelle Régionale; http://www.lmd.polytechnique.fr/chimere/; Schmidt et al., 2001) chemical mechanism was originally developed from an earlier version of the EMEP chemistry model (Simpson, 1992; Vautard et al., 2001), with special attention to low NO_x conditions and nighttime (NO_3) chemistry. The original extended version of the mechanism includes more than 300 chemical reactions of 80 gaseous species, whereas the reduced version (MELCHIOR2) treats 44 species and about 130 reactions. MELCHIOR2 has explicit oxidation schemes for methane, ethane, *n*-butane, ethene, propene and *o*-xylene. Biogenic compounds are represented by isoprene, α- and β-pinene, and lumped terpene, humulene and ocimene classes. Eight chemical operators (Carter, 1990; Aumont et al., 1996) are introduced in the reduced mechanism as surrogates for groups of reactive intermediates. In addition to the MELCHIOR2 gas-phase chemical mechanism, the CHIMERE modelling system also incorporates a sectional aerosol module with primary and secondary particles, multiphase sulphur and nitrogen chemistry, and the thermodynamic equilibrium scheme ISORROPIA (Nenes et al., 1998a, b).

The model has been applied e.g. to the simulation of air-pollution episodes at regional and urban scales and ozone-trend analyses (Beekmann and Vautard, 2009, and references therein). The CHIMERE/MELCHIOR modelling system has also been used in operational forecasting of pollutant levels over Western Europe for several years, and it has been extensively compared to observations (http://www.prevair.org; Honoré et al., 2008).

4.3.5 NWP-Chem (Enviro-HIRLAM)

The NWP-Chem scheme is an economical scheme designed at DMI for operational forecasting (Korsholm et al., 2008). It consists of the NWP-Chem-Gas gas-phase chemistry scheme and thermodynamic equilibrium model NWP-Chem-Liquid. The scheme includes 27 main reactions and describes the basic chemistry of the photo-oxidation of VOC to peroxy radicals, the most important NO_x reactions, the most important ozone formation reactions, sulphur (DMS = dimethyl sulfide is included) and isoprene chemistry (biogenic emissions of isoprene and other terpenes affects gas-phase chemistry – such as ozone – as well as aerosol formation). In the present version of NWP-Chem-Gas, the ordinary differential equations are solved using the quasi-steady-state approximation (Hesstvedt et al., 1978).

4.3.6 RADM2 (CAMx, CHEM, CMAQ, Enviro-HIRLAM, EURAD, OPANA, WRF-Chem) and RACM (Enviro-HIRLAM, EURAD, MOCAGE)

The second-generation Regional Acid Deposition Model (RADM2) gas-phase chemical mechanism (Stockwell et al., 1990) was developed from the earlier RADM mechanism (Stockwell, 1986). The emissions were aggregated into model species based on similarities in chemical reactivity, organic functional groups and the reactivity of the organic compounds with OH. The aggregation factors, rate parameters and product yields for the organic reactions were derived from the organic emissions aggregated into each model species (Middleton et al., 1990).

An evolution of RADM2-RADM, RACM (Regional Atmospheric Chemistry Mechanism), was proposed in (Stockwell et al., 1997). The reaction rate constants and product yields, as well as the cross sections and quantum yields for the photolysis were updated, and new condensed reaction mechanism was introduced for biogenic compounds. The mechanism was evaluated against data obtained from the University of California, Riverside, environmental chamber database (Carter et al., 1995).

4.3.7 SAPRC-99 (ALADIN-CAMx, CMAQ, FARM, OPANA)

The chemical mechanism developed at the Statewide Air Pollution Research Center in Riverside, California (SAPRC-99) is a detailed mechanism for the gas-phase atmospheric reactions of VOCs and oxides of nitrogen (NO_x) in urban and regional atmospheres (http://www.engr.ucr.edu/~carter/SAPRC99/). The scheme can be used in airshed models to determine absolute and relative ozone reactivities of many types of VOCs that can be emitted into the atmosphere, and for other control strategy and research applications. This mechanism represents a complete update of the Carter (1990) SAPRC-90 mechanism and incorporates recent reactivity data from a wide variety of VOCs. The mechanism has assignments for about 400 types of VOCs and can be used to estimate reactivities for about 550 VOC categories.

A condensed version of SAPRC-99 was developed for use in regional models. A unique feature of this mechanism is a computational system to estimate and generate complete reaction schemes for most non-aromatic hydrocarbons and oxygenates in the presence of NO_x, from which condensed mechanisms for the model can be derived. The mechanism was evaluated against the results of approximately 1700 environmental chamber experiments carried out at the University of California, Riverside, including experiments to test ozone reactivity predictions for over 80 types of VOCs. The mechanism was used to update the various ozone reactivity scales developed by Carter (1994), including the widely used Maximum Incremental Reactivity (MIR) scale. However, the reactivity estimates for many VOC classes are uncertain, which must be taken into account when using these data for regulatory applications. For this reason, uncertainty classifications have been assigned to all VOCs, and upper limit MIRs for VOCs with uncertain mechanisms are presented.

A new version of the SAPRC chemical mechanism called SAPRC-07 (not used at the moment in any of the discussed CWF models) is available online at http://www.engr.ucr.edu/~carter/SAPRC, including references to detailed description about improvements and new compounds in the new version.

4.3.8 UNI-OZONE (EMEP model, MATCH (EMEP-MSC-W))

The chemical scheme of the Unified EMEP Model (UNI-OZONE, http://www.emep.int/OpenSource/index.html; Simpson et al., 2003) contains full oxidant chemistry, gas and aqueous oxidation of SO_2 to sulphate, ammonium chemistry, nighttime production of HNO_3 and nitrate, coarse nitrate particle formation, as well as the advection of primary particles. Therefore, the scheme provides comprehensive chemistry for both photo-oxidant and acidification studies. The VOC scheme is lumped, with explicit oxidation mechanisms for methane, ethane, ethanol, *n*-butane, ethene, propene, *o*-xylene and isoprene. Since version 2.0, the module EQSAM (Metzger et al., 2002; Metzger, 2000) has been used in the Unified EMEP model to calculate the partitioning between gas and aerosol phase of HNO_3 and NO_3^- aerosol and NH_3 and NH_4^+ aerosol (Tarrasón et al., 2004).

The EMEP model is revised by the Executive Body for the Convention on Long-range Transboundary Air Pollution (LRTAP). In the 1990s, the EMEP models also became the reference atmospheric chemical transport model for use in the Integrated Assessment Models supporting the development of air-quality polices under the EU Commission. The chemical schemes of the EMEP model have been extensively intercompared with other atmospheric chemistry models (e.g. Kuhn et al., 1998; Andersson-Sköld and Simpson, 1999; Gross and Stockwell, 2003; Jimenez et al., 2003; Cuvelier et al., 2007; Vautard et al., 2007).

4.3.9 Synthesis and recommendations

All chemical sub-models discussed in this section are implemented at least in one CWF model. Several comparisons of the chemical schemes and also the modelling systems have been carried out and are documented in the literature. However, no one study covers all the schemes or sub-models discussed here, and both the objectives and the implementation of the intercomparisons differ greatly. Thus, it is not possible to rank the performance of the sub-models in relation to each other, based on the existing literature.

Table 5 provides a comprehensive overview of the intercomparisons between different chemical schemes, including some that were not part of this study. A common conclusion in several of the documented intercomparisons appears to be that most models are able to reproduce or predict the ozone concentrations fairly well, whereas they do not perform as well in simulating other compounds, such as NO_x and their reaction products (e.g. Kuhn et al., 1998; Gross and Stockwell, 2003; Jimenez et al., 2003; Vautard et al., 2007; Luecken et al., 2008). The skill of the models in simulating PM_{10} concentrations has also been poor or moderate (e.g. Vautard et al., 2007). A better understanding of the VOC oxidation mechanisms, especially because of their importance to secondary organic aerosol (SOA) formation (e.g. Kanakidou et al., 2005; Tunved et al., 2006) and the implementation of these processes in the CWF modelling systems also presents a formidable future challenge for chemical schemes.

The relative importance of the different components of chemical schemes (e.g. inorganic, organic, and aqueous phase chemistry) depends on the scientific aim and the applications of the CWF modelling system. Clearly, besides the structure of the chemical sub-module, the amount of available computer resources is another limiting factor for the accuracy of the concentration predictions. Available computer power may set an upper limit for the complexity of the chemical schemes that can be incorporated in the CWF modelling

Table 6. The treatments for aerosol particles in the CWF models: size distribution, chemical composition and aerosol microphysics.

Model	Size distribution representation			Chemical components in particle phase										Aerosol microphysics			
	Bulk	Modal	Sectional	Sea salt	Dust	Elem. C	Org. C	Sulfate	Nitrate	Ammonium	Water	Biog. SOA	Anthr. SOA	Nucl.	Cond.	Coag.	Dep.
ALADIN -CAMx	X		X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
CAMx -AMWFG	X		X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Enviro -HIRLAM		X		X	X			X						X	X	X	X
FARM		X		X	X	X	X	X	X	X	X	X	X	X	X	X	X
LOTOS -EUROS	X			X	X	X	X	X	X	X	X	X					X
MATCH	X			X	X	X	X	X	X	X	X						X
MM5 -CAMx	X		X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
MM5 -CHIMERE			X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
MM5 -CMAQ		X		X	X	X	X	X	X	X	X	X	X	X	X	X	X
NAME	X			X	X	X	X	X	X	X	X	X	X				X
OPANA		X		X	X	X	X	X	X	X	X	X	X	X	X	X	X
RCG	X			X	X	X	X	X	X	X	X	X	X				X
SILAM	X		X	X	X	X	X	X	X	X	X			X	X	X	X
SKIRON/ Dust	X				X												X
THOR	X			X													X
WRF -Chem		X(MADE)	X(MOSAIC)	X	X	X	X	X	X	X	X	X	X	X	X	X	X

systems. The aim for any CWF model is therefore to find the chemical sub-model with the best balance between scientific accuracy and computational efficiency.

4.4 Aerosol processes and microphysics

The demands for more accurate and detailed aerosol-size distribution, microphysics and chemistry description capability in atmospheric models have increased dramatically during recent years. On the one hand, this is because aerosols represent the largest uncertainty in global climate models when predicting radiative forcing (e.g. Solomon et al., 2007). On the other hand, and more relevantly to this review, particle size, composition and morphology are crucial to estimate lung penetration of aerosols and their health effects. This important motivation has resulted in a development and refinement of aerosol modules that are used in CWF models.

Atmospheric particulates have numerous sources, ranging from primary emissions (such as dust or pollen) to complicated aerosol formation processes involving gas-phase, liquid-phase and surface reactions (Seinfeld and Pandis, 1998). This presents several challenges to CWF models, especially as only a limited number of aerosol process submodels are generally available, and the state-of-the-art has not yet been established. As in the case of chemistry modules, there is no generally recognised single model that would be widely used by most of the CWF models.

The species covered by the various models include sea salt, dust, elemental carbon, organic carbon, sulfate, nitrate, ammonium, biogenic SOA and anthropogenic SOA (Table 6). Chemical weather models also commonly include only a fraction of the particulate matter components, leading to an underprediction of PM mass values. For example, in most cases, natural pollen is missing, suspended dust may be missing or not accurately evaluated, sea salt and emissions from wild-land fires may be missing, and secondary organic compounds are in many cases poorly represented. Due to the extreme complexity of this issue, easy-to-use semi-empirical methods have also been proposed for evaluating the long-range transported fraction of $PM_{2.5}$ (e.g. Kukkonen et al., 2008).

The different aerosol description options can be classified by (a) how the size distribution is represented and (b) what kind of aerosol microphysics is included in the modelling system. We classify the aerosol process methods according to the way they represent the size distribution. These can be grouped in three different categories: bulk schemes, modal schemes and sectional schemes. Some models have only one choice for the aerosol size distribution description, whereas others have several options.

One limitation to using detailed aerosol size distribution and composition descriptions arises from the lack of size- and composition-segregated emission data. The emission inventories are typically based on total mass only, and using a modal or sectional scheme requires assumptions about the emission size and chemical composition distributions.

The state-of-art at present consists of a size-resolved sectional representation for the aerosol size distribution with

equilibrium chemistry partitioning packages for both inorganics and organics. In addition, all major microphysical processes (nucleation, coagulation, condensation, wet and dry deposition) are included, as well as schemes for biogenic and anthropogenic SOA formation. Table 6 lists different characteristics of each of the various aerosol modules.

4.4.1 Bulk schemes

In *bulk schemes*, typically the total mass of suspended particles (TSP) or the mass in a certain size interval, or several non-interacting intervals, is modelled. The intervals are typically one or some combination of PM_1, $PM_{2.5}$, PM_{10} and TSP. Such an approach is computationally efficient, but naturally has severe limitations when size-dependent processes are important.

To estimate the health effects of the respirable particles better, the focus of both measurements and modelling has gradually moved from PM_{10} and TSP to $PM_{2.5}$ and PM_1. Since the lung penetration function and the health effects are dependent in a complicated manner on both the size and chemical composition, bulk schemes will likely be replaced gradually by the more resolved (but computationally more expensive) modal and sectional schemes (Zhang et al., 1999).

4.4.2 Modal schemes

In *modal schemes* (Whitby and McMurry, 1997), the aerosol size distribution is represented by a small number of modes (size categories), the properties of which are modelled as functions of time and location. This typically involves a predescribed assumption (e.g. log-normal) of the functional form of the modes. This approach is computationally more expensive than bulk methods, but less resource-consuming than sectional methods. For this reason, such schemes have been quite common in regional and global models. The performance of modal schemes is limited when new-particle formation is important. One additional challenge in modal schemes is due to pre-assumed size- and composition ranges of the modes, resulting in a need for redistributions of the particles between modes (e.g. Vignati et al., 2004).

4.4.3 Sectional schemes

In *sectional schemes* (Jacobson, 2005), the continuous size distribution is replaced by a large number of discrete bins (i.e. the size distribution is approximated by a histogram). The model has equations for the particle concentration (number or mass) and chemical composition that are solved for each bin. The sectional scheme is the most flexible and accurate one, but it is computationally the most expensive. With increasing computing power and memory, more CWF models are adopting sectional schemes as their choice for aerosol size distribution representation. A major challenge for CWFs using sectional aerosol representations is improving the quality and level of detail of the emission inventories to match the sophistication of the rest of the model, which is a concern regarding both size and chemical-composition distributions of the emissions.

4.4.4 Aerosol microphysics

The main microphysical processes affecting the aerosol size distribution are nucleation, condensation/evaporation, coagulation and deposition (Seinfeld and Pandis, 1998). Nucleation, or the formation of new particles through a gas-to-particle phase change, has been observed to occur throughout the atmosphere (e.g. Kulmala et al., 2004) and is an important particle source, especially in the nucleation and Aitken mode size ranges. Clearly, particle growth by condensation does not change the number concentration, but alters particle size and the mass concentration. Atmospheric coagulation is typically a process between small nucleation or Aitken mode particles and larger accumulation-mode or coarse-mode particles. Coagulation does not change the mass concentration, but decreases the number concentration of particles.

In bulk schemes, deposition is considered without an accurate way to describe its dependence upon particle size. Condensation/evaporation is usually treated by assuming equilibrium between the gas and particle phases, by using a chemical equilibrium thermodynamics scheme, such as ISORROPIA (Nenes et al., 1998a) or the Equilibrium Simplified Aerosol Module (EQSAM; Metzger et al., 2002). In sectional and modal schemes, all the above-mentioned microphysical processes can be adequately described, which is important especially when detailed information is desired on the particle number concentration distribution or the chemical composition distribution as a function of size. A size-resolved model is also crucial because different microphysical processes depend on different aerosol properties which cannot be taken into account using a bulk scheme representation.

4.5 Deposition

Dry and wet deposition are processes that remove pollutants from the atmosphere. Not only are accurate schemes required for producing realistic concentrations of pollutants in the atmosphere, but deposited pollutants can affect soil and vegetation (e.g. acidification) and water bodies (e.g. eutrophication). The spatial distributions of wet and dry deposition are therefore commonly assessed in the various long-term environmental assessment programmes (e.g. EMEP). Uncertainties in modelling deposition, however, can also limit short-term forecasts of ground-level pollutant concentrations. For example, sensitivity tests by Wesely and Hicks (2000) showed that daytime ozone concentration could increase by about 20 % when dry deposition is not acting.

References and brief characterizations of the dry and wet parameterization schemes used in the CWF models considered are summarized in Table 7.

Table 7. References or brief characterizations of the treatments for dry and wet deposition in the CWF models.

Model name	Canopy Resistance	Aerosol settling velocity and deposition	In-cloud scavenging	Below-cloud scavenging
CAMx	Wesely (1989)	Slinn and Slinn (1980)	Different scavenging coefficients for gases and aerosols following Seinfeld and Pandis (1998); precipitating water, snow and ice considered	Different scavenging coefficients for gases and aerosols following Seinfeld and Pandis (1998); precipitating water, snow and ice considered
Enviro-HIRLAM	Wesely (1989) and Binkowski (1999)	Näslund and Thaning (1991)	Scavenging coefficients for gases following Seinfeld and Pandis (1998); in-cloud scavenging of aerosols dependent on the aerosol radius and rain rate (Baklanov and Sørensen, 2001)	Scavenging coefficients for gases following Seinfeld and Pandis (1998); below-cloud scavenging of aerosols dependent on the aerosol radius and rain rate (Baklanov and Sørensen, 2001)
EURAD-RIU	Walcek et al. (1986)	Size dependent resistance model and gravitational settling depending on the three aerosol lognormal modes	Different scavenging coefficients based on Henrys law equilibria for each specie; aerosol mode dependent scavenging (Binkowski, 1999)	Different scavenging coefficients based on Henrys law equilibria for each specie; aerosol mode dependent scavenging (Binkowski, 1999)
FARM	Wesely (1989)	Seinfeld and Pandis (1998); Binkowski and Shankar (1995)	Gas scavenging as in EMEP Unified model (Simpson et al., 2003)	Gas scavenging as in EMEP Unified model (Simpson et al., 2003); aerosols following Scott (1978)
LOTOS-EUROS	Erisman et al. (1994)	Erisman and Draaijers (1995)	Neglected at start of paper writing, now via scavenging coefficients	Gases: scavenging rates depending on Henry's law constant and precipitation intensity (Simpson et al., 2003); aerosols following Scott (1978)
MATCH	Erisman et al. (1994) Bartnicki et al. (2001)	Seinfeld and Pandis (1998)	Ozone, H_2O_2 and SO_2 in-cloud scavenging is calculated by assuming Henry's law equilibrium in the clouds; for sulfate particles, in-cloud scavenging is assumed to be 100 % effective	For sulfate particles Berge (1993); neglected for ozone, H_2O_2 and SO_2; for other species proportional to the precipitation intensity and a species-specific scavenging coefficient
CHIMERE	Erisman et al. (1994)	Seinfeld and Pandis (1998); Zhang et al. (2001); Giorgi (1986); Peters and Eiden (1992)	Dissolution of gases in cloud droplets (Seinfeld and Pandis, 1998); aerosol nucleation (Tsyro, 2002; Guelle et al., 1998)	Dissolution of gases in precipitating drops (Mircea and Stefan, 1998); scavenging by raining drops (Loosmore and Cederwall, 2004)
CMAQ	RADM (Wesely, 1989); M3Dry (Pleim et al., 2001)	Binkowski and Shankar (1995)	Wet deposition algorithms taken from RADM (Chang et al., 1987); wet deposition of chemical species depending on precipitation rate and cloud water concentration (Roselle and Binkowski, 1999); accumulation and coarse mode aerosols completely absorbed by cloud and rain water, Aitken mode aerosols slowly absorbed into cloud and rain water	Wet deposition algorithms taken from RADM (Chang et al., 1987); wet deposition of chemical species depending on precipitation rate and cloud water concentration (Roselle and Binkowski, 1999); accumulation and coarse mode aerosols completely absorbed by cloud and rain water, Aitken mode aerosols slowly absorbed into cloud and rain water
MOCAGE	ISBA (Interface Soil Biosphere Atmosphere) scheme (Michou et al., 2004)	Nho-Kim et al. (2004)	Convective (Mari et al., 2000) and stratiform precipitation (Giorgi and Chameides, 1986)	Giorgi and Chameides (1986)
NAME	Resistance analogy incorporating canopy resistance	Resistance analogy; particles removed by sedimentation and impaction with the surface	Rain and snow scavenging coefficients for large-scale and convective precipitation (Maryon et al., 1996)	Rain and snow scavenging coefficients for large-scale and convective precipitation (Maryon et al., 1996)
OPANA	Wesely (1989)	Binkowski and Shankar (1995)	Wet deposition algorithms taken from RADM (Chang et al., 1987); wet deposition of chemical species depending on precipitation rate and cloud water concentration (Roselle and Binkowski, 1999); accumulation and coarse mode aerosols completely absorbed by cloud and rain water, Aitken mode aerosols slowly absorbed into cloud and rain water	Wet deposition algorithms taken from RADM (Chang et al., 1987); wet deposition of chemical species depending on precipitation rate and cloud water concentration (Roselle and Binkowski, 1999); accumulation and coarse mode aerosols completely absorbed by cloud and rain water, Aitken mode aerosols slowly absorbed into cloud and rain water
RCG	Erisman et al. (1994)	From Stoke's law (Pleim et al., 1984)	Neglected	Species dependent scavenging coefficients for gases from Henry constant and precipitation rate (Seinfeld and Pandis, 1998); scavenging coefficient identical coefficients for all particles
SILAM	Hicks et al. (1987); Lindfors et al. (1991)	From Stoke's law	Water and snow scavenging from large-scale and convective precipitation (Smith and Clark, 1989; Jylhä, 1991)	Water and snow scavenging from large-scale and convective precipitation (Smith and Clark, 1989; Jylhä, 1991)

Table 7. Continued.

Model name	Canopy Resistance	Aerosol settling velocity and deposition	In-cloud scavenging	Below-cloud scavenging
SKIRON/ Dust	–	Slinn and Slinn (1980), Kumar et al. (1996)	Constant scavenging coefficient (Seinfeld and Pandis, 1998)	Constant scavenging coefficient (Seinfeld and Pandis, 1998)
THOR	Wesely and Hicks (1977)	Gravitational settling velocity given by Stokes equation (Hanna et al., 1991)	Rain and snow scavenging coefficients for large-scale and convective precipitation (Maryon et al., 1996)	Rain and snow scavenging coefficients for large-scale and convective precipitation (Maryon et al., 1996)
WRF/ CHEM	Wesely (1989) and Erisman et al. (1994)	Slinn and Slinn (1980), Pleim et al. (1984)	In-cloud wet removal of aerosol particles involves removal of the cloud-borne aerosol particles collected by rain, graupel and snow, using the same first-order rate that cloud water is converted to precipitation. For trace gases, the same removal rate is applied to the fraction of each gas that is dissolved in cloud water	Below-cloud wet removal of aerosol particles by impaction scavenging via convective brownian diffusion and gravitational or inertial capture. Irreversible uptake of H_2SO_4, HNO_3, HCl, NH_3 and simultaneous reactive uptake of SO_2, H_2O_2. (Easter, 2004)

4.5.1 Dry deposition

Dry deposition is governed by the turbulent and molecular diffusion of pollutants in the atmosphere, and the gravitational settling. The turbulent and molecular diffusion depends upon the characteristics of the surface, vegetation, and the physical and chemical properties of the depositing species (e.g. the solubility and chemical reactivity for gases and the size distribution and chemical composition for particles). Gravitational settling needs to be accounted for for coarse particles. For example, Seinfeld and Pandis (1998) and Sportisse (2007) provide more comprehensive descriptions of deposition.

Therefore, a successful dry deposition scheme should be capable of reproducing both the boundary-layer turbulent fluxes and the interaction between the pollutant and the surface. The choice of parameterization is conditioned by the meteorological model, which provides the surface-layer turbulence, by the surface and soil characteristics and by input data availability. For example, in regional models, bulk schemes for vegetation canopies (often called big-leaf schemes) are generally preferred to so-called multi-layer canopy models. The latter could be considered more suitable to describe deposition processes within tall canopies, but their use may be hindered by the lack of input data to describe the vertical structure of vegetation.

Dry deposition is commonly formulated in Eulerian models as a boundary condition at the ground surface for the vertical diffusion term of the pollutant transport equation. In this term, a species-dependent vertical concentration flux is the product of a deposition velocity V_d and the surface concentration. In state-of-the-art CWF models, the mathematical treatments of the dry deposition for gases and aerosols are usually based on the resistance analogy, where the inverse deposition velocity is the sum of three different resistances in series ($V_d^{-1} = R_a + R_b + R_c$): the aerodynamic resistance R_a, due to turbulent diffusion, the quasi-laminar layer resistance R_b, due to molecular diffusion, and the canopy resistance R_c, due to the capture of pollutants by the surface (e.g. Wesely, 1989; Seinfeld and Pandis, 1998). Differences in modelling dry deposition among various CWF models arise from different ways to estimate the resistance terms, but also from the way the CWF models are interfaced with the meteorological models. The aerodynamic and quasi-laminar resistances are a function of the atmospheric stability and friction velocity (u_*), which depend on the coupling strategy chosen for each modelling system. On-line coupled models and some off-line coupled models use parameters (e.g. surface momentum flux) provided by the meteorological model, whereas other off-line models prefer to re-estimate them through diagnostic parameterizations usually based on similarity theory. These different approaches can cause differences in the predicted deposition, even if we consider models implementing the exact same parameterization.

The dry deposition schemes in the CWF models in this study are largely similar. The aerodynamic resistance R_a and the quasi-laminar sub-layer resistance R_b are parameterized in terms of the friction velocity, surface roughness and molecular diffusivity of species (Wesely and Hicks, 1977; Walcek et al., 1986; Hicks et al., 1987; Chang et al., 1987, 1990; Wesely, 1989). This approach is adopted by all the models considered in this paper. The only different approach is implemented by CMAQ in its second dry deposition scheme (M3Dry), where R_a is computed coherently with MM5 land-surface model (Byun and Ching, 1999; Pleim and Xiu, 1995) from the surface heat flux and the difference in virtual potential temperature between the air and the ground.

Greater differences among the CWF models occur for the parameterization implemented for the surface resistance R_c (Table 7). Usually, the surface resistance is expressed as a set of parallel resistances associated with leaf stomata, leaf cuticles, other canopy structures (e.g. bark, stems), and surface soil and water. Over land, R_c can be expressed

as the sum of foliar (R_{cf}) and ground (R_{cg}) resistances ($1/R_c = 1/R_{cf} + 1/R_{cg}$), and foliar resistance is subdivided in stomatal (R_{st}) and non-stomatal or cuticle (R_{cut}) resistances ($1/R_{cf} = 1/R_{st} + 1/R_{cut}$). Many different approaches have been developed for the calculation of stomatal resistance, varying from a simple function of solar radiation and temperature (Wesely, 1989), a big-leaf approach taking into account air temperature and humidity, together with leaf-area index and canopy wetness (Hicks et al., 1987), to a multilayer leaf-resistance model (Baldocchi et al., 1987). For non-stomatal resistance, a constant value is often chosen, depending on season and land type (e.g. Wesely, 1989 and Zhang et al., 2002). Other models use meteorological variables such as u_* and canopy height as scaling parameters to characterize in-canopy aerodynamic resistance, and relative humidity to describe the cuticle resistance (Erisman et al., 1994). A more detailed parameterization for cuticles taking into account meteorological and season-dependent vegetation parameters has been proposed by Zhang et al. (2003). The influence of chemical-physical properties of depositing gaseous species has been analyzed by Zhang et al. (2002), who scaled cuticle and ground resistances of the species considered by their CTM to O_3 and SO_2 resistances on the basis of published dry deposition measurements and of the evaluation of their aqueous solubility and oxidizing capacity. The parameterizations implemented within each model for canopy resistance are summarized in Table 7.

The dry deposition velocity of particles V_d can be written as $V_d = V_s + (R_a + R_b + R_a R_b V_s)^{-1}$, where V_s is the settling velocity. This formula is derived assuming that particle settling operates in parallel with the three resistances already introduced for gases operating in series. This approach is implemented in almost all the CWF models (Table 7).

Although the differences in these mathematical treatments may seem small, they can nevertheless result in substantial differences to the model predictions. For example, Sportisse (2007) showed that the implementation of a different mass-conserving formula, expressed as $V_d = V_s(1 - \exp(-V_s(R_a + R_b))^{-1}$ (Venkatram and Pleim, 1999), can reduce coarse-particle deposition velocities in low-wind conditions by up to 20 %. Published comparisons of deposition velocities obtained by different models applied on the same areas showed uncertainties of ± 30 % (Wesely and Hicks, 2000). Timin et al. (2007) performed a sensitivity analysis of CMAQ surface concentrations to the dry deposition scheme, showing that the simpler scheme available in CMAQ (based on Wesely, 1989) produces lower deposition velocities for all the species and increases in ozone 8-h average concentrations up to 10–20 ppb with respect to the more up-to-date M3Dry scheme (Pleim et al., 2001). Dry deposition parameterization has been identified as one of the main causes of differences between their CMAQ and CAMx simulations.

In their summary of dry deposition, Wesely and Hicks (2000) found that resistance schemes are quite reliable in daytime conditions over flat terrain, but are less reliable for mountainous areas and during nighttime stable conditions. The reasons are that the parameterizations of aerodynamic resistance, usually based on surface-layer similarity theory, do not provide an accurate evaluation of turbulent mixing during stable stratification and in complex terrain.

Additional uncertainties reside in the subgrid variation of surface and land-use features, where horizontal advection effects are not considered in summing the contributions from different patches with different surface effects. For homogeneous atmospheric and surface conditions, improper definition of surface features, e.g. vegetation and soil moisture, can result in large differences between modeled and measured deposition. Zhang et al. (2003) showed that a detailed description of cuticles and soil resistances can improve the description of daily variation and maximum value of deposition velocity for wet canopies. In such conditions, stomatal uptake is not important, due to stomata blocking by waterdrops and to the presence of very weak solar radiation. Zhang et al. (2002) analyzed the variation of V_d values for the 31 species modeled by their CTM AURAMS (A Unified (multiple-pollutant) size- and chemical composition-resolved, episodic, Regional Air-quality Modelling System) considering their dependence on land-cover, season and daytime. They computed values varying within an order of magnitude for vegetated surfaces, ranging between 0.3 and $5\,\mathrm{cm\,s^{-1}}$ over deciduous broadleaf forest during summer daytime conditions, and higher values for species with very high solubility and oxidizing capacity.

The evaluation of dry deposition models is quite difficult due to the lack of direct measurements of deposition fluxes. As a consequence, very few evaluation studies including extensive comparison with observations are available in the literature. Petroff et al. (2008) recently compared the performances of selected analytical and differential dry deposition models for aerosols versus measurements over grass and forest. Analytical models rely on parameterizations of different complexity, as those previously mentioned for the different resistances. Differential models solve the differential transport equations for the different chemical species within the canopy layer, and require as input vertical profiles of parameters describing vegetation features, as the leaf-area density.

Differences in the deposition velocity of up to one order of magnitude have been obtained for fine particles (Petroff et al., 2008). Analytical models (Slinn, 1982; Zhang et al., 2001) displayed small variations in the deposition velocity when applied to grass and forest. In contrast, differential models (Davidson et al., 1982; Wiman and Agren, 1985) exhibited large differences in deposition velocity, but showed a strong dependence on parameters describing canopy geometry and aerodynamics, such as the leaf-area index, obstacle size, roughness length and displacement height – properties that can be difficult to determine for regional-model applications.

4.5.2 Wet deposition

Wet deposition refers to scavenging of contaminants and their transport to the earth's surface by atmospheric hydrometeors and is usually subdivided into in-cloud scavenging (rainout) and below-cloud scavenging (washout). Although dry deposition is introduced in Eulerian numerical models as a lower boundary condition in flux form, wet deposition is described as a depletion term within the transport-diffusion equation for pollutant concentration and can be parameterized by $\mathrm{d}C/\mathrm{d}t = -\Lambda C$, where C is the substance concentration and Λ is the scavenging coefficient (s^{-1}). The scavenging coefficient is different from zero where precipitation occurs and in the presence of condensation (clouds or fog). The existing computational schemes for the scavenging coefficient range from simple functions of rain rate and cloud-water content, to complex models describing the system of physical, microphysical and chemical processes that characterize the interaction of gases and aerosols with cloud condensate and precipitation (e.g. Seinfeld and Pandis, 1998; Sportisse, 2007).

Simple parameterizations could potentially be sufficient for CWF models, especially for off-line coupled models, which have no access to the full meteorological model microphysics. For a reliable short-term estimate of near-ground air-pollutant concentrations, below-cloud scavenging is expected to dominate, at least in areas characterized by relevant local and regional emissions – in other words, where short-range transport dominates over long-range sources (e.g. in continental and Mediterranean Europe). Neglecting in-cloud scavenging should underestimate the mass of deposited pollutant, but have only a weak effect on surface concentrations. Moreover, cloud-aerosol interactions can modify precipitation rate and its spatial distribution, and therefore indirectly influence near-surface scavenging. However, these phenomena can be described only by on-line coupled CWFs that can implement cloud-pollutant interactions and can take into account feedback effects of air pollution on meteorology. Wet deposition schemes vary much more than the dry deposition schemes for the operational CWF models in this article. For example, LOTOS-EUROS, MATCH, FARM and RCG use simple parameterizations of scavenging rates that are similar to those implemented in the EMEP Unified model (Simpson et al., 2003). These depend on Henry's law constant, rain rate and cloud-water mixing ratio for gases, and, on particle size, precipitation intensity and raindrop fall speed for aerosols. The possible release of scavenged gases and aerosols due to cloud- or rain-water evaporation is not taken into account by the latter parameterization. In contrast, NAME, SILAM and THOR use scavenging coefficients depending upon cloud type (convective vs. stratiform) and precipitation type (rain vs. snow). Other models, such as CHIMERE and Enviro-HIRLAM, use more complex in-cloud and below-cloud scavenging parametrizations, whereas LOTOS-EUROS and RCG neglect in-cloud scavenging.

Even though rain has a polydisperse distribution of drop size and pollutant scavenging is dependent upon the fall velocity of the droplets, expressing the scavenging coefficients as a simple function of rain rate, assuming a monodisperse raindrop distribution, appears to be justified as long as a representative droplet diameter is chosen (Wang et al., 2010), although this choice depends upon the properties of the pollutant (Mircea and Stefan, 1998; Andronache, 2003; Sportisse, 2007). The main uncertainty of this simplified approach, implemented in almost all the CWF models considered here, is how the rainfall intensity is determined and how it varies over the specified time interval from which it is output from the NWP model. Although wet deposition seems weakly related to drop size, it is much more strongly dependent upon aerosol size. For example, Baklanov and Sørensen (2001) and Andronache (2003) showed that below-cloud scavenging was dependent upon aerosol size distribution, being important for very small ($<0.01\,\mu\mathrm{m}$) and coarse ($>2\,\mu\mathrm{m}$) particles. Therefore, boundary-layer aerosol-size distribution can be modified by precipitation, with quick removal of coarse particles. A proper description of aerosol size distribution within precipitation scavenging parameterizations is therefore required to estimate size-resolved particulate matter (PM) concentrations.

Comparing the results from wet deposition schemes implemented inside different models is difficult because of the complexity of the CWF models (e.g. spatial and temporal differences between forecasted cloud and precipitation, aerosol size and composition). Textor et al. (2007) compared results from 16 global models participating in the AeroCom project (Schulz et al., 2009) and found a large variability in the ability of models to handle wet deposition. They had difficulty in identifying the reasons for the main differences (e.g. the models did not provide the same indication about the type of rain; convective or stratiform precipitation) that was most efficient in removing aerosols from the atmosphere.

Wang et al. (2010) recently performed a systematic examination of the uncertainties on below-cloud size-resolved scavenging coefficients for particles, considering both analytical and empirical parameterizations. The largest uncertainties were associated with specification of the raindrop-particle collection efficiency. The use of different formulas can result in differences in Λ values up to two orders of magnitude for particles in the range of 0.01–3 µm. The use of various raindrop number distributions can cause differences between a factor three and five for all particle sizes, whereas the uncertainty caused by different raindrop settling velocity formulations is smaller than a factor of two. Comparison with field measurements showed that most size-resolved Λ parameterizations underestimate experimental values up to two orders of magnitude for particles smaller than 3 µm. This difference is not justified by the combined effect of the previously mentioned uncertainties, but is attributed to additional known physical processes (e.g. turbulent transport and mixing) that can influence field data, but are not taken into

account by parameterizations.

The predicted size-resolved particle concentrations using different Λ parameterizations can differ by up to a factor of two for particles smaller than 0.01 µm and by a factor of more than ten for particles larger than 3 µm after 2–5 mm of rain. The predicted bulk mass concentrations can differ by a factor of two between theoretical and empirical parameterizations after 2–5 mm of moderate intensity rainfall.

The major source of uncertainty for estimates of wet deposition is the rain rate, because biases in the precipitation predictions greatly affect the wet deposition estimates. Furthermore, the accurate estimation of precipitation amounts and location is still one the most difficult and challenging tasks for meteorological models, especially for convective systems.

4.6 Natural emissions

Emissions can be broadly classified into natural and anthropogenic ones. Natural emissions is a wide term that includes different compounds (e.g. NO_x, SO_2, NH_3, PM, Non-Methane Volatile Organic Compounds (NMVOCs), CH_4 and CO) emitted from sources such as vegetation, soils, animals, wetlands, sea salt, primary biological aerosol particles, wind-blown dust, volcanoes, lightning, forest fires, etc. Anthropogenic emission inventories are significant components in our growing effort to understand the impact of human activity on air quality, particularly in the large urban areas (Markakis et al., 2010). They represent important input data to CTMs (Russell and Dennis, 2000).

In the last years, CTMs have improved greatly and consequently more detailed and accurate anthropogenic emission data are needed. Modern anthropogenic emission inventories should have high temporal and spatial resolutions, include a large variety of anthropogenic emission sources and account for many different chemical compounds emitted. Probably the most commonly used anthropogenic emission inventory for Europe is that of the EMEP (http://www.ceip.at/). Anthropogenic emission inventories that can be used for operational air-quality forecasting over Europe have also been developed within the framework of EU projects, such as GEMS (Visschedijk et al., 2007) and MACC (Visschedijk et al., 2010). Anthropogenic emission inventories are not addressed in detail in this study, but some challenges in their development are discussed in Sect. 7.1.

Air pollutants from natural sources play a prominent role in the physics and chemistry of the atmosphere and also contribute to the ambient air concentrations of air pollutants (e.g. O_3, PM, SOA; Seinfeld and Pandis, 1998). For example, the organic compounds released in the atmosphere by vegetation, collectively referred to as Biogenic Non-Methane Volatile Organic Compounds (BNMVOCs), contribute to the formation of O_3 (Curci et al., 2009; Wang et al., 2008; Bell and Ellis, 2004) and SOA (Kleindienst et al., 2007; Kanakidou et al., 2005). Curci et al. (2009) simulated an average 5 % increase in summer daily ozone maxima over Europe due to BNMVOCs emissions with peaks over Portugal and the Mediterranean Region (+15 %). BNMVOCs suppress the concentrations of the hydroxyl radical (OH), enhance the production of peroxy (HO_2 and RO_2) radicals and generate organic nitrates that can sequester NO_x and allow long-range transport of reactive nitrogen (Fehsenfeld et al., 1992).

The salt flux from the sea surface is an important factor in the formation of cloud condensation nuclei (CCN) in the marine boundary layer. The salt flux also affects sea surface-atmosphere exchange and heterogeneous chemistry, including the oxidation of SO_2 and NO_2 in the marine boundary layer (Foltescu et al., 2005; Pryor et al., 2001). In Europe, the contribution of mineral dust to PM_{10} concentrations varies from 10 to more than 30 % depending on location and season (Putaud et al., 2004) and, in the United States, the fraction of mineral dust found in $PM_{2.5}$ exceeds 10 % in most areas and reaches 50 % in dry areas (Malm et al., 2004; Park et al., 2010).

Volcanoes release considerable fluxes of gases and particles to the atmosphere, both during eruptions and by long-term noneruptive degassing. Water, carbon dioxide, and sulphur species represent by far the predominant component of volcanic gases. In Europe, significant volcanic emissions have been to date limited to Italy and Iceland. An extensive compilation of available, measured volcanic sulphur fluxes has been carried out for the Global Emissions Inventory Activity (GEIA) (Andres and Kasgnoc, 1998). The dataset contains volcanic SO_2 emissions averaged over 25 yr from the early 1970s to 1997. It includes average SO_2 emissions from 49 continuously emitting volcanoes (four located in Europe: Etna, Stromboli, Vulcano and Kverkfjoll) and maximum SO_2 emissions from 25 sporadically emitting volcanoes (none located in Europe).

The gaseous and particulate natural emissions accounted for in the CWF systems, as well as their calculation methodologies, are presented in Table 8.

4.6.1 Natural gaseous emissions

In most cases, the methodologies for the quantification of natural emissions require input data such as emission potentials based on measurements, meteorological data and land-use data (e.g. land cover, leaf-area index) derived from satellite observations. The estimated natural emissions are gridded data and have to be speciated according to chemical mechanisms used by the photochemical grid models. On the European scale, there are some studies focusing on estimating natural emissions and their impact on air quality (Simpson et al., 1999; NATAIR, 2007; Curci et al., 2009). However, the uncertainties in natural emissions remain large (larger than those of anthropogenic emissions).

Almost all of the 18 CWF models use biogenic emissions (e.g. isoprene, monoterpenes, other volatile organic compounds) in the forecast runs. Biogenic emissions are

Table 8a. The gaseous natural emissions accounted for in the CWF models, as well as their calculation methodologies. Only part of the selected 18 models have been presented in the table, as some of the models do not include gaseous natural emissions.

Model name	Gaseous emissions									
	Vegetation		Soil		Volcanoes		Oceans		Animals (wild and domestic)	
	Component	Method	Component	Method	Component	Method	Component	Method	Component	Method
ALADIN-CAMx	Isoprene, 2-Methyl-3-buten-2-ol, Methanol, Ethene, Propene, Ethanol, Acetone, Hexanal, Hexenol, Hexenylacetate, Formaldehyde, Acetaldehyde, Butene, Ethane, Formic acid, Acetic acid, Butenone, other reactive VOCs and Monoterpenes	SMOKE emission model (Houyoux and Vukovich, 1999) using the Biogenic Emissions Inventory System (BEIS3) (http://www.epa.gov/asmdnerl/biogen.html); Guenther et al. (1994, 2000)	Nitric oxide, Carbon monoxide	SMOKE emission model (Houyoux and Vukovich, 1999) using the BEIS3 mechanism (http://www.epa.gov/asmdnerl/biogen.html); Guenther et al. (2000)	Not included	Not included	Not included	Not included	Not included	Not included
CAMx-AMWFG	Isoprene	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org)	Ammonia	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org)	Sulphur dioxide	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org). Continuously erupting volcanoes	Ammonia	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org)	Ammonia	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org)
ENVIRO-HIRLAM	Isoprene, monoterpene, methylbutenol, methanol, nitric oxide, 3-carene, limonene, myrcene, ocimene, alpha-pinene, beta-pinene, sabinene	MEGAN, v.2.1 (Guenther et al., 2006)	Not included	Not included	Not included	Not included	DMS, Ammonia, Carbon monoxide	MOZARD	Ammonia	EDGAR
FARM	Isoprene, Monterpenes	Guenther et al. (1993, 1995)	Nitric oxide	Willliams et al. (1992)	Not included	Not included	Not included	Not included	Not included	Not included
LOTOS-EUROS	Isoprene, Monterpenes	Guenther et al. (1993, 1995)	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included
MATCH	Isoprene	Simpson et al. (1995) (E-94 methodology); Guenther et al. (1993)	Not included	Not included	Sulphur dioxide, sulphate	EMEP expert emission inventory (http://www.emep.int/)	Not included	Not included	Ammonia	EMEP expert emission inventory (http://www.emep.int/)
MM5-CAMx	Isoprene, Monoterpenes, other VOCs	Emission model developed by the Aristotle University of Thessaloniki and the National and Kapodistrian University of Athens (Poupkou et al., 2010; Symeonidis et al., 2008; Guenther et al., 1995)	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included
MM5-CHIMERE	Isoprene, *A*-pinene, *B*-pinene, Limonene, Ocimene	MEGAN emission model (Guenther et al., 2006)	Nitric oxide	Stohl et al. (1996)	Not included	Not included	Not included	Not included	Not included	Not included
MM5-CMAQ	Isoprene, Monoterpenes	BIOEMI emission model developed by the Technical University of Madrid (Guenther et al., 1993, 1995; Schoenemeyer et al., 1997; Steinbrecher, 1997)	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included

Table 8a. Continued.

Model name	Gaseous emissions									
	Vegetation		Soil		Volcanoes		Oceans		Animals (wild and domestic)	
	Component	Method	Component	Method	Component	Method	Component	Method	Component	Method
MOCAGE	Isoprene, Monoterpenes, other VOCs	Guenther et al. (1993, 1995)	Nitrogen oxides, Carbon monoxide	Emmons and Lamarque (NCAR), personnal communication (2006)	Sulphur dioxide	Andres and Kasgnoc (1998)	Carbon monox-ide, Methane	Emmons and Lamarque (NCAR), personnal communication (2006)	Not included	Not included
NAME	No natural emissions									
OPANA	Isoprene, Monterpenes	BIOEMI emission model developed by the Technical University of Madrid (Guenther et al., 1995; Schoenemeyer et al., 1997; Steinbrecher, 1997)	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included
RCG	Isoprene, Monoterpenes, other VOCs	Simpson et al. (1995) (E-94 methodology); Guenther et al. (1993); CORINAIR (CORe INventory of AIR emissions) emission handbook	Nitric oxide	Simpson et al. (1995)	Not included	Not included	Not included	Not included	Not included	Not included
SILAM	Isoprene, Monterpenes	Guenther et al. (1993, 1995)	Ammonia	EMEP/GEMS/ GEIA/RETRO emission invemtories	Sulphur oxides	EMEP/GEMS/ GEIA/RETRO emission invemtories	Not included	Not included	Ammonia	EMEP/GEMS/ GEIA/RETRO emission invemtories
SKIRON/ Dust	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included	Not included
THOR	Isoprene	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org)	Nitrogen oxides	Global Emissions Inventory Activity (GEIA) database (www.geiacenter.org)	Not included	Not included	Not included	Not included	Ammonia	Global Emissions Inventory Activity (GEIA) database (www. geiacenter.org)
WRF-Chem	Isoprene, Monoterpenes, other VOCs	Biogenic emission module based on Guenther et al. (1993, 1994), Hahn et al. (1994), Simpson et al. (1995), Schoenemeyer et al. (1997) or the Biogenic Emissions Inventory System (BEIS3) (Vukovich and Pierce, 2002)	Nitric oxide	Simpson et al. (1995)	Not included	Not included	Not included	Not included	Not included	Not included

Table 8b. The particulate emissions accounted for in the CWF models, as well as their calculation methodologies.

Model name	Particulate natural emissions		
	Dust Methodology	Sea salt Methodology	Pollen Methodology
ALADIN-CAMx	Not included	Not included	Not included
CAMx-AMWFG	Desert dust fluxes from the SKIRON/Dust modelling system	Module developed by the AM&WF Group at the National and Kapodistrian University of Athens (de Leeuw et al., 2000; Gong et al., 2002; Gong, 2003; Zhang et al., 2005b; Shankar et al., 2005; Astitha and Kallos, 2009)	Not included
ENVIRO-HIRLAM	Zakey et al. (2006)	Zakey et al. (2008)	Birch pollen emission module developed by the Danish Meteorological Institute and the Finish Meteorological Institute (Mahura et al., 2009)
FARM	Vautard et al. (2005)	Zhang et al. (2005)	Not included
LOTOS-EUROS	Not included	Monahan et al. (1986); TNO (2005)	Not included
MATCH	Only anthropogenic: Andersson et al. (2009)	Foltescu et al. (2005)	Not included
MM5-CAMx	Not included	Not included	Not included
MM5-CHIMERE	Vautard et al. (2005), Marticorena and Bergametti (1995), Menut et al. (2007)	Monahan et al. (1986)	Not included
MM5-CMAQ	Not included	Not included	Not included
MOCAGE	Martet et al. (2009)	Gong et al. (1997)	Not included
NAME	No natural emissions		
OPANA	Not included	Not included	Not included
RCG	Loosmore and Hunt (2000), Claiborn et al. (1998)	Gong et al. (1997)	Not included
SILAM	Not included	Module developed in the Finish Meteorological Institute based on Monahan et al. (1986) and Martensson et al. (2003)	Modules developed in the Finish Meteorological Institute for birch and grass pollen (Sofiev et al., 2006b)
SKIRON/ Dust	Dust module developed by the AM&WF Group at the National and Kapodistrian University of Athens (Marticorena and Bergametti, 1995; Nickovic et al., 2001; Zender et al., 2003; Pérez et al., 2006)	Not included	Not included
THOR	Not included	Not included	Not included
WRF-Chem	Not included	Not included	Not included

mostly calculated from emission models (MEGAN – Model of Emissions of Gases and Aerosols from Nature), BEIS3, AUTH-NKUA – Aristotle University of Thessaloniki-National and Kapodistrian University of Athens model, BIOEMI – BIOgenic EMIssion model) and modules or, in a few cases, they are taken from existing databases. The algorithms that are usually applied are those introduced by Guenther et al. (1993, 1994, 1995), according to which isoprene emissions are temperature and light dependent whereas monoterpenes and other VOC emissions are temperature dependent. Additional processes relevant with the emissions of biogenic compounds are described by some of the emission models. For example, BEIS3 provides species-specific, seasonally-dependent biogenic emission factors and leaf-area index for each land-use type, adjusting the isoprene emissions for the effects of the Photosynthetically Active Radiation penetrating through the leaf canopy. Another example is the AUTH-NKUA model, which accounts for the light dependency of monoterpenes emissions from some vegetation species.

MEGAN describes the variation of biogenic emissions as a function of numerous environmental variables and factors (e.g. temperature, light, humidity, wind within the canopy, leaf-area index, leaf age, soil moisture), whereas it also accounts for the losses and productions in the canopy. Emissions from soils (mainly nitric oxide as a function of soil temperature, land use and fertilizer input) are used as input data to only some of the CWF models (ALADIN-CAMx, CAMx-AMWFG, FARM, MM5-CHIMERE, MOCAGE, RCG, SILAM, THOR, WRF-Chem). Other gaseous natural emissions (e.g. volcanoes, oceans, animals) are hardly accounted for in chemical weather forecast models (only in CAMx-AMWFG, MOCAGE, SILAM and THOR). Lightning emissions of NO_x from the GEIA database are used in the operational runs of THOR.

4.6.2 Natural particulate matter emissions

Some particulates occur naturally, originating from volcanoes, dust storms, forest and grassland fires, living vegetation, and sea spray. In this section, we focus on primary aerosol emissions, especially dust and sea-salt particles, which constitute the largest contribution to total aerosol mass. Other particulates are formed by way of the transformations of pollutants such as sulfur dioxide, nitrogen oxides and ammonia into sulfates, nitrates and ammonium, respectively (secondary aerosol). Many volatile organic compounds are converted to oxidized organic species with low volatility, thus becoming a component of ambient aerosol.

FARM and MM5-CHIMERE use a simplified bulk scheme for the calculation of mineral dust emissions as in Vautard et al. (2005) (cf. Table 8). Vautard et al. (2005) also propose a simplified scheme to calculate the emissions that depends upon turbulence near the ground, assuming that the resuspension of material that is available on the ground can explain the missing parts of the PM_{10} average load. The desert dust emission fluxes mainly depend on wind velocity and the surface features (Marticorena and Bergametti, 1995). The emission modules account for the effects of the soil size distribution, surface roughness and soil moisture. The dust module developed by the AM&WF (Atmospheric Modelling and Weather Forecasting Group, School of Physics, University of Athens, Athens, Greece) Group is used by SKIRON/Dust and CAMx-AMWFG. The current model version incorporates state-of-the-art parameterizations of all the major phases of the atmospheric dust life cycle such as production, diffusion, advection, and removal, including the effects of the particle size distribution on aerosol dispersion and deposition. Different size bins can be considered with diameters ranging from 0.1–10 µm following a log-normal distribution (Zender et al., 2003). During the model run, the prognostic atmospheric and hydrological conditions are used to calculate the effective rates of the injected dust concentration based on the viscous/turbulent mixing, shear-free convection diffusion and soil moisture. The RCG model uses the methods by Loosmore and Hunt (2000) and Claiborn (1998) to calculate the resuspension of dust. MOCAGE has been coupled with a module of dynamic source of dust emissions using a size-resolved (bin) approach (Martet et al., 2009).

Sea-spray droplets come in three varieties: film, jet, and spume. Film and jet droplets derive from one process: air bubbles bursting at the sea surface. When a bubble rises to the surface, its film-thin top eventually ruptures and ejects tens to hundreds of film droplets with radii ranging roughly from 0.5 to 5 µm. After the bubble bursts, it collapses and, in so doing, shoots up a jet of water from its bottom. Because of velocity differences along this jet, it soon breaks up into a few jet droplets with radii typically from 3 to 50 µm, depending on the size of the bubble. Spume droplets derive from another process: the wind tears them off the wave crests. Estimating the sea-salt emissions, and hence the amount of local marine aerosol, requires knowing the rate at which spray droplets of any given size are produced at the sea surface (i.e. the sea-spray generation function). LOTOS-EUROS, SILAM and MM5-CHIMERE quantify the bubble- and spume-production mechanisms using the approach of Monahan et al. (1986).

In SILAM, the aerosol-size distribution is extended to submicrometer particles according to Martensson et al. (2003); a detailed description has been presented by Sofiev et al. (2011b). RCG and CAMx-AMWFG also simulate the processes of sea-salt aerosol generation, diffusive transport, transformation, and removal as a function of particle size (Gong et al., 1997). FARM and CAMx-AMWFG implemented the methods proposed by Zhang et al. (2005), which corrects sea-salt-emission particle-size distributions according to local relative humidity.

In comparison with the main anthropogenic emission sources, the pollen particles emitted depends on meteorological conditions. The emission modules for pollen therefore

should include treatments for the effects of the accumulated heat, start and end dates of the pollinating season, mean climatological rate of release, correction functions related to wind, temperature, humidity, and precipitation, and diurnal cycle of the pollen production. Pollen emissions are used as input emission data only in SILAM and Enviro-HIRLAM forecast runs. For this reason, emission modules have been developed by FMI and DMI. The most difficult problems in pollen-dispersion are to evaluate the emission flux of grains and their time evolution. For example, trial forecasts during spring 2004 with SILAM used a "climatologic" emission term, which was based on the results of long-term mean observed birch flowering dates (Sofiev et al., 2006a). The system is based on the European flowering start and duration maps from the International Phenological Garden Project (IPG, 2004). The maps were compiled by Rötzer and Chmielewski (2001) using multilinear regression analysis of phenological observations in Europe over 35 yr (1961–1998).

Mahura et al. (2009) investigate the patterns of birch pollen counts over a diurnal cycle and propose a parameterization that is useful for inclusion into operational and research short- and long-term modelling with Enviro-HIRLAM for birch pollen atmospheric transport and deposition at different spatial scales. The evaluation of patterns of diurnal cycles on monthly and interannual bases has been done based on analysis of a 26-yr time series of birch pollen counts from the Danish pollen measurement site in Copenhagen. The suggested parameterization, based on a simple trigonometric function, includes dependencies on the time of birch pollen maximum and minimum occurrence on a diurnal cycle, averaged concentration at the end of the previous day, and time shift.

Some models also account for biomass burning and wildland fires. For example, THOR uses the emission dataset of the EU project REanalysis of the TROpospheric chemical composition over the past 40 yr (RETRO). In contrast, for SILAM, Sofiev et al. (2009) investigated the potential of two remotely-sensed wildland fire characteristics (4 µm brightness temperature anomaly – TA, and fire radiative power – FRP) for the needs of operational chemical transport modelling and short-term forecasting of atmospheric composition and air quality. The core of the methodology is based on empirical emission factors that are used to convert the observed temperature anomalies and fire radiative powers into emission fluxes. A new generation fire assimilation system is presented, which evaluates the daily, global emission fluxes of primary particulate matter from wildland fires. The predicted emissions in Europe are subsequently scaled to other pollutants using emission factors from the literature and submitted to the chemical transport model SILAM for diagnostic assessment and forecasting of the atmospheric composition.

4.7 Horizontal and vertical grid spacing

In this article, we distinguish between *grid spacing* and *resolution* of the CWF models (Pielke, 1991, 2001; Laprise, 1992; Grasso, 2000a, b). Grid spacing for Eulerian models can be simply defined as the distance between numerical grid points; however, resolution can in many cases be an ambiguous or poorly defined concept. Resolution commonly refers to the smallest spatial or temporal scale, on which various phenomena can be resolved by the model or modelling system. However, for instance the minimum distance scale on which phenomena can be resolved, may vary from two to ten times the grid spacing. The resolution (or grid spacing) can also be different for the emission data, meteorological data, land-use data, other input data, the computational grid of the model, the receptor grid of the model, and other data. Thus, the meaning of the overall resolution of the modelling system, and how it has been evaluated for a specified phenomenon, may not always be clear. Therefore, whenever possible we prefer to use the more precise term grid spacing.

Lagrangian models also face a similar problem. For instance, the effective grid spacing for NAME is determined by the meteorological fields that the model uses and by the grid spacing of the emissions. In general, for both Lagrangian and Eulerian models, the effective resolution of the modelling system is no better than the coarsest of the grids that are employed by the NWP model, the emissions processing model, and the CTM.

A summary of the computational grid spacings and coordinate systems of the different models appear in Table 9. Clearly, CWF models can be implemented with various horizontal and vertical grid spacings, depending upon the atmospheric scales to be modeled. Computational time limitations of having forecasts appear in real-time during operational forecasts restrict the domain size and grid spacing. Modelling on the continental, regional, and background urban scales necessarily require different horizontal grid spacings. For example, a CWF model for forecasting regional or municipal air quality requires small horizontal grid spacing (e.g. of the order of from 1 to 20 km), but does not necessarily require model levels in the stratosphere. In contrast, continental-scale models typically have 10–50-km horizontal grid spacing and should include treatments for the entire troposphere and the lower stratosphere for many applications. Clearly, the selection of the appropriate grid spacing depends upon the details of the modelling system and the particular application.

Most of the models described in this paper use multiple grids that may have different horizontal grid spacings for the meteorological and air-quality components (for the off-line models). In that way, the CWF models may cover the continental and regional scale across Europe and, with the finer grids (sometimes nested), they may focus on a more detailed forecast of a specific region. For instance, operational modelling on the European scale in 2010 featured horizontal

Table 9. Details on the grid spacings and coordinate systems of the different CWF models.

Model name	Coordinate system	Horizontal grid spacing	Vertical grid spacing
ALADIN-CAMx	Arakawa C grid	28.9 km for the mother grid (Central Europe) 9.63 km for the inner modelling domain (Austria and surroundings)	15 vertical layers (30 m to 2.5 km). The highest grid spacing (about 30 m) is achieved in the lowest 5 levels (up to about 350 m)
CAMx-AMWFG	Arakawa C grid	$0.24^\circ \times 0.24^\circ$ ($\sim$ 24 km) The area covered is the Mediterranean region, Europe (up to 55° N), North and Central Africa, Turkey and part of the Arabian Peninsula	22 layers up to 8 km with variable spacing First layer from the ground at 50 m
ENVIRO-HIRLAM	Arakawa C grid	5 km (pollen forecast)	Hybrid terrain-following sigma and pressure coordinate system 40 layers, grid spacing from 30 m to 500 m
EURAD-RIU		Europe: 125 km; Central Europe: 25 km, German States: 5 km	23 layers from 40 m to 2000 m at top (100 hPa)
FARM	UTM (Universal Transverse Mercatore); polar stereographic	12 km (Italian Peninsula)	Terrain following coordinates with variable vertical spacing up to 10 km
LOTOS-EUROS		$0.5^\circ \times 0.25^\circ$ (Europe) ($\sim 25 \times 25$ km) $0.25^\circ \times 0.125^\circ$ (Netherlands) ($\sim 12 \times 12$ km) $0.125^\circ \times 0.0625^\circ$ (Netherlands) ($\sim 6 \times 6$ km)	Dynamic mixing layer approach 4 layers (surface layer of 25 m, mixing height layer and 2 reservoir layers up to 3.5 or optionally 5 km)
MATCH		44 km (MATCH-HIRLAM) (Europe) 0.5° and 0.2° ($\sim$ 50 km and 20 km, respectively) (MATCH-ECMWF) (Europe)	Usually depending on met. model. At present for HIRLAM: domain height $\sim$ 8 km, lowest level at $\sim$ 60 m
MM5-CAMx		30 km for Europe (mother grid) 10 km for the Balkan Peninsula and 2 km for Athens (nested grids)	CAMx: 15 vertical layers, 1st layer height 20 m, top at 7 km. MM5: 29 vertical sigma-levels, top at 100 mbar
MM5-CHIMERE		50 km (Western Europe) 10 km (Portugal)	In the vertical there are 8 layers up to 500 hPa with the surface layer located at 50 m
MM5-CMAQ		50 km (Europe) 27 km (Iberian Peninsula)	15 layers up to 100 hPa
MOCAGE		PREVAIR: 2° ($\sim$ 200 km) (global); 0.5° ($\sim$ 50 km) (Europe and Mediterraean area); 0.1° ($\sim$ 10 km) (France) GEMS, MACC: 2° (global); 0.2° (Europe) Currently testing: 0.025° (France)	Hybrid (sigma, P) coordinate system with currently 60 levels from the surface up to 1 or 0.1 hPa (7–8 levels in the PBL with a first layer of 20 to 40 m)
NAME		No intrinsic grid. The Met Office Unified: globally at 40 km resolution and in a European limited area configuration at 12 km	Continuously variable
OPANA	UTM	5 km (coarse grid, Madrid) 1 km (nested grid, Madrid)	Terrain following coordinates with 15 layers up to 6 km. Surface layer at 20 m
RCG		25 km	5 layers, surface layer of 25 m, 2 layers above surface layer and mixing height and 2 reservoir layers
SILAM		$0.2^\circ \times 0.2^\circ$ ($\sim$ 20 km) (Europe) 5 km (Northern Europe)	Multi-vertical approach with the meteorology-resolving grid corresponding to the tropospheric part of the IFS vertical: hybrid levels. The chemical transformations and vertical fluxes are computed on the basis of thick staggered layers

Table 9. Continued.

Model name	Coordinate system	Horizontal grid spacing	Vertical grid spacing
SKIRON/Dust	Polar stereoraphic Arakawa E-grid	$0.05 \times 0.05^\circ$ (~ 5 km) 2 grids: one for the Mediterranean Region and Europe and one extended to the North Atlantic Region	Eta step-mountain vertical coordinate system with 38 vertical levels from the surface up to 22 km
THOR		DEHM mother domain: 150×150 km (hemispheric) DEHM first nest: $50\,\text{km} \times 50\,\text{km}$ (Europe) DEHM second nest: 16.67×16.67 km UBM: $1\,\text{km} \times 1\,\text{km}$ (urban) OSPM: 0.001 km (street pollution)	DEHM: 20 layers up to $\sim$15 km, lowest model layer 50 m
WRF-Chem		50 km (Europe)	The vertical structure has 12 layers in sigma coordinates with the top pressure at 100 hPa
WRF/CMAQ	Arakawa C	12 km	Terrain-following hydrostatic pressure coordinates. 22 layers extending from the surface to 100 hPa are interpolated from the 60 WRF layers

grid spacings in the range of 20 km (SILAM), 25 km (CAMx-AMWFG, RCG, SKIRON/Dust, EURAD-RIU), 30 km (ALADIN-CAMx, MM5-CAMx), 44 km (MATCH-HIRLAM) and 50 km (LOTOS-EUROS, MATCH-ECMWF, MM5-CHIMERE, MM5-CMAQ, MOCAGE, THOR, WRF-CHEM).

For the finer-grid forecast simulations, the variety of horizontal grid spacing in 2010 ranged from 2 km (MM5-CAMx for Athens area, MOCAGE over France at 2.5 km) to 27 km (MM5-CMAQ for the Iberian Peninsula) for the three-dimensional Eulerian models. Many models use 10–12-km horizontal grid spacing for their finer grids (MM5-CAMx for the Balkan region, MM5-CHIMERE for Portugal, MOCAGE for France and WRF/CMAQ) and 5 km for Northern Europe (Enviro-HIRLAM, EURAD-RIU and SILAM). The other applied horizontal grid spacing is 9.6 km for ALADIN-CAMx covering Austria, 12 km for FARM (Italian Peninsula), 25×12 km for LOTOS-EUROS (covering the Netherlands) and 17 km for THOR.

As well as the grid spacing, another parameter that differs among models and applications is the selection of the coordinate system. Horizontal spatial coordinates may be expressed in polar coordinates on a sphere, Cartesian coordinates on a plane, or one of several projections of a sphere onto a plane. Curvilinear coordinates may be used in both polar and planar instances, where the model refers to a pseudo-longitude and latitude, that is then mapped to geographic longitude and latitude (following the curved surface of the earth).

Following the Cartesian map projections (fixed physical distance coordinates on a flat plane), a number of models included in this paper use the Lambert Conic Conformal coordinate system (ALADIN-CAMx, MM5-CAMx, MM5-CMAQ, WRF-Chem and WRF-CMAQ) for the forecasting applications. Another Cartesian map projection is the Universal Transverse Mercator (FARM). A rotated longitude-latitude grid is used by Enviro-HIRLAM model, and a curvilinear geodetic latitude-longitude projection is used by CAMx-AMWFG and LOTOS-EUROS. In all geographic projections, the surface of the Earth is distorted because the Earth's actual shape is irregular. Nevertheless, all projections produce similar results and most of the models allow the user to select the map projection among different options.

The models also have different vertical coordinate systems describing how the grid levels are separated in the vertical: height, terrain-following σ, pressure, and step-mountain coordinates. LOTOS-EURO uses a fifth system, a dynamic mixing-layer coordinate system with four layers (a surface layer of 25 m, mixing-layer height, and two reservoir layers up to 3.5 or 5 km). RCG also uses a dynamic mixing-layer coordinate system with a surface level, 25-m surface layer, mixing layer and two reservoir layers up to 4 km. Uniformity appears on the selection of the surface layer, where most of the models use 20–50 m above the surface as the first model level. Also, most of the models are focused on the troposphere with the top layers located at 2.5–15 km, with two exceptions at 22 km (SKIRON/Dust) and 1 hPa ($\sim$ 50 km, MOCAGE).

5 Sensitivity analysis and evaluation of CWF models

In this article, we use the term "model evaluation against data" or in abbreviated form "model evaluation" to refer to a systematic comparison of model predictions and observations. We avoid the term "validation", as, strictly speaking,

Table 10. A summary description of the evaluation of each CWF model.

Model name	How model was evaluated	References (up to 4)	Quantities evaluated
ALADIN-CAMx	Citydelta project: aimed to explore the changes in urban air quality predicted by different chemistry-transport dispersion models in response to changes in urban emissions. Model provided good performances for ozone (both on average and for extreme values). Acceptable results have been obtained for PM_{10} yearly means. ESCOMPTE project: the model has highlighted quite good performance for both ozone and NO_2	- Hirtl et al. (2007) - Baumann-Stanzer et al. (2005) - Vautard et al. (2007)	O_3, O_x, PM_{10}, NO_2
CAMx-AMWFG	The evaluation of the model performance on the known gas and aerosol species has been included in several publications worldwide. The model intercomparison has been done against in-situ measurements for the species concentration and against AERONET data for the dust optical depth used for the calculation of the photolysis rates	- Astitha et al. (2006) - Astitha et al. (2007) - Astitha and Kallos (2009) - Astitha et al. (2010)	The evaluation of the model performance on the known gas and aerosol species such as ozone, NO_x, sulfates, nitrates etc.
Enviro-HIRLAM	Evaluation against field experiments of ETEX and MEGAPOLI, and Chernobyl measurements. Meteorology and air-quality forecasts evaluated against Paris surface observation network for specific episodes. Model intercomparison: participant in EU MEGAPOLI project. Needs further evaluation over long-term periods	- Chenevez et al. (2004) - Korsholm et al. (2009) - Korsholm (2009) - Mahura et al. (2008)	Transport and scavenging processes have been evaluated using ETEX and Chernobyl observations; meteorology (with feedbacks), surface O_3, NO_x, SO_2, PM using Paris monitoring and MEGAPOLI campaign data
EURAD-RIU	GEMS: evaluation against measurements and other air-quality forecast models. Participation of the COST 728 model intercomparison for the winter 2003 case	- Hass et al. (1997) - Jakobs et al. (2002) - Schlünzen and Fock (2010)	O_3, PM_{10}, NO_2,
FARM	On single model components and against monitoring data in real applications. Long-term model intercomparison exercise over Po Valley (Northern Italy), carried out by Regional Environmental Protection Agencies. Ongoing long-term model intercomparison exercise over Po Valley (Northern Italy), carried out by Regional Environmental Protection Agencies	- Silibello et al. (2008) - Schlünzen and Fock (2010) - Calori et al. (2010) - Gariazzo et al. (2007)	O_3, NO_2, NO_x, PM_{10}
LOTOS-EUROS	Evaluation with groundbased measurements. EURODELTA: a regional-scale model intercomparison to analyse the responses of different CTMs to emission changes/scenarios	- Schaap et al. (2008)	O_3, NO_2, NO, NH_3, SO_4, SO_2 and NH_4. Secondary organic aerosols, sea salt, and heavy metal concentrations
MATCH	Eurodelta: evaluation of seven regional air-quality models and their ensemble for Europe and Mics Asia – Model intercomparison study for Southern and Eastern Asia, Phase 1 and 2	- Carmichael et al. (2002) - Carmichael et al. (2008b)	Evaluated reference dataset: chemistry and transport including SO_2, NO_2, NO_x, NH_3, HNO_3, O_3, CO, CF_6, ^{137}Cs, seasalt, (CF_6 during the ETEX-experiment and ^{137}Cs for the Chernobyl accident). Model intercomparison: including SO_2, NO, NO_2, NO_x, NH_3, HNO_3, O_3, HCHO
MM5-CAMx	Within the European project GEMs, the air quality forecast has been operationally evaluated against surface measurements in Europe (rural stations of EMEP, urban stations of AIRBASE in Athens, Greece) and compared with the forecasts from other models (e.g. CHIMERE, EMEP, EURAD etc.) and the European ensemble forecast. Tropospheric columns of NO_2 and O_3 have been compared with satellite data. There has been also upper air evaluation with WOUDC sites measurements	- Huijnen et al. (2010) - Kioutsioukis et al. (2009) - Kioutsioukis et al. (2010) - Poupkou et al. (2008)	Ozone, nitrogen dioxide, sulphur dioxide, carbon monoxide and particulate matter (PM_{10})

Table 10. Continued.

Model name	How model was evaluated	References (up to 4)	Quantities evaluated
MM5-CHIMERE	MM5: evaluated in many model intercomparison studies, particularly compared to RAMS. CHIMERE: analysis evaluation was performed through studies published in more than 30 perr reviewed papers. For the forecast, the evaluation of the model is updated daily on the PREVAIR web site with correlations scores compared to hourly surface emasurements. Model intercomparison: City-Delta, Euro-Delta, Esquif, escompte. Numerous projects, described on the CHIMERE web site	- Hara et al. (2005) - O'Neil et al. (2005) - Menut et al. (2005) - Vautard et al. (2007)	Dispersion simulations of O_3, O_x, PM_{10}
MM5/WRF-CMAQ	The model is used for several experiments and compared the results with the observational data. Results show correlation coefficients between 0.5 to 0.9 for Ozone concentrations for one year hourly concentrations (8760 data)	- Meng et al. (2007) - Vijayaraghavan et al. (2007) - San José et al. (2007) - Appel et al. (2009)	Analysis of the amount of air pollutant concentrations due to the industrial plant emissions. Evaluation of the potential impact of an incinerator. Modelling the transport and transformation of mercury. Performance of the model system is compared with the existing measurements of a total of 22 PCB congeners and the 17 most toxic PCDD/F congeners
MOCAGE	Meteorological forcings from MOCAGE are provided by numerical weather prediction suites at Météo-France (ARPEGE, ALADIN) and ECMWF (IFS), with operational skill score evaluations. MOCAGE has been evaluated against observations in the context of a range of field campaigns and international exercises (ESQUIF, ESCOMPTE, City-Delta, ICARTT-ITOP,...), with over 40 publications in the international refereed literature. Evaluation range from the global scale (including the stratosphere) to the regional/local scale for gases and primary aerosol species. Continuous operational skill score monitoring for regulatory species is performed at Météo-France and INERIS in the context of PREVAIR (Rouil et al., 2009)	- Josse et al. (2004) - Dufour et al. (2004) - Bousserez et al. (2007) - Ménégoz et al. (2009)	Transport and scavenging processes have been evaluated using ETEX and Rn/Pb observations; surface O_3, NO_x, SO_2, HNO_3, PAN using routine surface observation and campaign data; deposition of ozone using ESCOMPTE data; global tropospheric and stratospheric distributions of Ozone, CO, NO_2, N_2O using a range of satellite data products; aerosol was evaluated using surface PM observations, Lidar and AERONET data, as well as campaign data
NAME	Evaluation against field experiments including ETEX, and Kincaid. Air-quality forecasts evaluated against UK surface obs network. Model intercomparison: participant in EU ENSEMBLE project	- Webster and Thomson (2002) - Ryall and Maryon (1998) - Simmonds et al. (1996)	Plume rise scheme is evaluated against Kincaid dataset. NAME model predictions are compared against ETEX. Model has been evaluated against observations of a number of trace gases. Intercomparisons amongst European models used to simulate foot and mouth disease spread
OPANA	Evaluated at the end of EMMA project (CGXIII, 1996–98). The system has been tested again many data during the last 10 years. Model intercomparison: the model has been tested and compared with observational data in every air quality impact assessment (callibration phase) and in every real-time air-quality forecasting system developed for urban and/or industrial areas because the system is callibrated with one-year air-quality monitoring data in the subjected area and surroundings	- San José et al. (2005)	O_3, NO_2, CO, SO_2, PM_{10}, $PM_{2.5}$, Cadmium, Arsenic, Nickel, Lead and Benzo(a)pyrene

Table 10. Continued.

Model name	How model was evaluated	References (up to 4)	Quantities evaluated
RCG	PM_{10}-measurements done in and aroud the Greater Berlin Area have been used to evaluate RCG on different scales. EuroDelta model intercomparison for Europe. EUROTRAC – GLOREAM: the focus is primarily on model performance for aerosol components in conjunction with the EMEP observational data that has been extended using Dutch and German special observation sites	- Beekmann et al. (2007) - van Loon et al. (2004) - Hass et al. (2003)	PM_{10} (EC, OC, inorganic aerosols, metals), sulphate, nitrate, ammonium, elemental carbon, wind-blown-dust events
SILAM	Regular emergency-type evaluations whenever possible. Operational evaluation of the air-quality forecasts using present-week observations over Finland. European-scale re-analysis for 2000–2003. Emergency-type model intercomparisons within EU-ENSEMBLE and follow-up projects, NKS-MetNet network, etc. Air-quality intercomparison projects are on-going within the scope of COST-728, EU-GEMS, and ESA-PROMOTE	- Sofiev et al. (2006c) - Galmarini et al. (2004a) - Huijnen et al. (2010)	Air-quality forecasts. Individual model units were compared against analytical solutions, chemical scheme tested as a box model, etc.
SKIRON/Dust	evaluation of the model SKIRON/Dust have been performed from AM&WFG during several projects (SKIRON, MEDUSE and ADIOS). Also, the modelling system has been used by other Universities and Institutes world-wide. Model intercomparison has been performed against measurements and observations	- Kallos et al. (2007) - Kallos et al. (2009) - Astitha and Kallos (2009) - Spyrou et al. (2010)	Intercomparison against in-situ measurements of dust and PM concentration, remote measurements of aerosol optical depth from satellites or radars

Table 10. Continued.

Model name	How model was evaluated	Evaluation level	References (up to 4)	Quantities evaluated
THOR	EuroDelta experiment: long-term ozone simulations from seven regional air-quality models were intercompared and compared to ozone measurements. Evaluation for two cities in Denmark with Urban Backround Model, BUM, and Operational Street Pollution Model, OSPM, included to THOR system	Level 2	- Brandt et al. (2001a)	Performance of the air pollution models BUM and OSP for NO_x, O_3, NO, NO_2
WRF-Chem	WRF-Chem-MADRID has beeb evaluated with Satellite and Surface Measurements	Level 2	- Zhang et al. (2005a)	The simulated concentrations of gas and aerosol species (e.g., O_3, SO_2, NO_x, and $PM_{2.5}$) and aerosol optical properties (e.g., aerosol optical depth, single scattering albedo, aerosol direct radiative) are being compared against available observational data

a complete validation of any atmospheric model is not possible. We use the term "verification" to refer to the testing of the fidelity of the computer code to the model equations and principles.

To develop and improve CWF models, different approaches can be employed to evaluate their skill and usefulness. In this section, we explore some of those approaches through sensitivity analysis (Sect. 5.1), individual model evaluation studies (Sect. 5.2), and multiple-model evaluation studies (Sect. 5.3). Also, the evaluation practices of each of the 18 models were reviewed and presented in Table 10.

5.1 Sensitivity analysis

The dynamical evolution of numerically simulated chemical weather is highly dependent upon uncertainties in the model structure (e.g. physical and computational parameterizations), as well as uncertainties in the input data (e.g. initial and boundary conditions, emissions). Such model behaviour can be investigated through sensitivity analysis, which seeks to determine the variation in model output as a function of variations in input variables and parameters (forward sensitivity analysis: applied to source-oriented modelling) or

the variation in model inputs resulting from variations in the model output (adjoint sensitivity analysis: applied to receptor-oriented modelling). Sensitivity analysis could provide information on the input factors that are mainly responsible for the output uncertainties (forward) or the input factors that are mostly responsible for the discrepancy between the model output and the observations (adjoint). Observational data is therefore not used in forward sensitivity analysis – it is only required in adjoint sensitivity analysis.

Uncertainty and sensitivity analysis assist in understanding the relative importance of different processes in the atmosphere and in quantifying the impact (either singular or with interactions) of uncertain inputs (e.g. data, parameterizations) in the results. Sensitivity information from CWF models can be useful for various purposes, such as the design of optimal pollution control strategies, the estimation of model parameters, research for the improvement of forecast skill, the evaluation of the role of processes (e.g. emissions, chemical kinetics, boundary conditions, parameterizations of vertical diffusion, etc.), source apportionment, and data assimilation.

The quantitative apportionment of the variation in the modelled concentrations to different sources of variation is accomplished through either the *statistical* or the *deterministic* sensitivity analysis approach. In the *statistical* approach (e.g. Hanna et al., 2001; Tang et al., 2010), the model is executed several times, each time with slightly perturbed inputs and the sensitivity is estimated from the statistical properties of the multiple output variability. In the *deterministic* approach (e.g. Dunker et al., 2002; Hakami et al., 2003, 2006; Zhang et al., 2005b; Napelenok et al., 2006; Koo et al., 2007), the model output equations are differentiated with respect to its inputs, and the sensitivity is calculated simultaneously with the concentration fields through an auxiliary set of equations.

Deterministic sensitivity analysis techniques propagate the derivatives either forward or backward along the model trajectories. In the forward method (e.g. tangent linear model, direct decoupled method or DDM), the uncertain inputs are perturbed and these perturbations are propagated forward through the modelling domain at future times, providing sensitivity information at all receptors with respect to a few uncertain parameters (*forward sensitivities*). Technically, this can be accomplished using either additional differential equations (e.g. Dunker et al., 2002) or by inserting additional lines of code in the model that calculate the gradient of the output function at each point (e.g. Carmichael et al., 1997). In the backward method (adjoint model), the perturbation is made at the receptor end and is propagated backward in time and space, providing sensitivity information about specific receptors with respect to all sources and parameters (*adjoint sensitivities*) (e.g. Sandu et al., 2005). Implementation of adjoint sensitivities in CWF models is increasing, mainly because of their application in chemical data assimilation.

Of the 18 models reviewed in this article, the following sensitivity analysis modules are available. Forward sensitivity analysis modules (DDM) are included in CAMx and CMAQ. The adjoint of CMAQ is available, although the WRF-Chem adjoint is under development. The EURAD-IM (Inverse Model) is capable of both forward and backward sensitivity tests, and the MOCAGE-PALM system is a useful platform for sensitivity studies in chemical data assimilation. Published sensitivity analysis studies performed with these CWF models are listed in Table 13.

Statistical sensitivity analysis techniques do not require any model modification because they rely on multiple model simulations with different combinations of the uncertain inputs. The most common and representative methods are reviewed by Saltelli et al. (2000). The statistical approach has limited applications in three-dimensional CWF models, principally due to its high computational requirements and also its restrictions on the statistical distribution of uncertain inputs. Ensemble prediction appears to be a better framework to deal with those restrictions, as it can provide information both about the forecast uncertainty and the *ensemble sensitivity*, using a mixture of formal statistical treatments and an informal treatment on some parts of the modelling cascade.

5.2 Evaluation of individual models against data

Before a CWF model can be used as an operational tool, model users should ensure that all the stages of the evaluation have been critically performed. Although evaluating a CWF model under all circumstances and for all applications is not possible, evaluation for each specific application is more feasible. Therefore, proper assessment involves determining whether the model is properly simulating the spatial and temporal features on the scales resolved by the model and whether the physical and chemical processes are simulated correctly in the model, leading to proper model response to changes in meteorology and emissions.

The main goal of model evaluation is to demonstrate that the model is making reasonable predictions when compared with observations, taking into account the adequacy and accuracy of the science represented in the model for the purposes for which the model is applied (e.g. Britter et al., 1995). Such evaluation exercises are usually based on the analysis of the systematic biases and errors in model outcomes, together with correlation measures, but they should also indicate sensitivities and uncertainties in the atmospheric processes simulated within the model. The results of these exercises should lead to new directions in model development and improvement, as well as point to the need for additional measurements.

Several studies have discussed the evaluation of CWF models and the importance of improved characterization of model uncertainties (e.g. Chang and Hanna, 2004; Fox, 1984; Demerjian, 1985; Borrego et al., 2008, Dennis et al., 2010; Schlünzen and Fock, 2010), as well as

suggestions for model evaluation methods (e.g. Venkatram, 1979, 1988; Weil et al., 1992; Dabberdt et al., 2004). Recently the Air-Quality Modelling Evaluation International Initiative (http://aqmeii.jrc.ec.europa.eu/) based its activities on a model evaluation framework that considers four main evaluation types: (i) operational (ii) diagnostic, (iii) dynamic and (iv) probabilistic.

In the first step, commonly referred to as operational evaluation, model predictions are compared to observed data, and some statistical measures are computed to gauge overall model performance. This evaluation against data determines how accurately the model predicts the real world from the perspective of the intended uses of the model. Schlünzen and Fock (2010) present an overview of the most common statistical parameters used to indicate the ability of the model to predict the tendency of observed values, errors on the simulation of average and peak observed values, and the type of errors (systematic or unsystematic). According to Weil et al. (1992) and Hanna et al. (1993), in the early 1990s, three performance measures were regularly applied to CWF model evaluation: mean bias, root mean square error, and correlation. Currently, a more extensive collection of statistical measures is commonly used. In particular, operational evaluation should include a calculation of the statistical confidence levels.

Clearly, there are several categories of measurements: ongoing routine network measurements and short-term, higher-resource campaign measurements. These different types of measurements have different uses, advantages and limitations, and consequently, different degrees of uncertainty.

In the next steps (diagnostic and dynamic evaluation), the objective is to address whether the predicted concentrations stem from correctly modelled or incorrectly modelled processes, whether they be physical or chemical. This evaluation step determines whether the model implementation accurately represents the developer's conceptual description of the model and the solution to the simulation. These evaluation methods can cover a wide variety of evaluation studies that consider the physical, chemical, meteorological and emission processes.

Finally, model evaluation can include probabilistic evaluation that attempts to capture the uncertainty or confidence in model results for chemical weather forecasting applications. Many methods exist to estimate the uncertainty: ensemble runs (i.e. multiple runs with different configurations of the same model), direct calculation of variances in predicted concentrations, Monte Carlo runs, and analytical error-propagation methods for simple-model algorithms. This probabilistic model evaluation should allow quantification of the confidence in model-predicted values and determination of how observed concentrations compare within an range of uncertainty for model predictions. Sensitivity tests (Sect. 5.1) are one of the most common and traditional ways to ascertain whether inputs have a notable influence on model performance issues.

Because the majority of the model systems described in this article are based on a NWP model and on a chemistry-transport model, a two-stage evaluation procedure (in which the weather forecast is evaluated independently of the chemistry model) is often the common model evaluation strategy. As a large experience with NWP evaluation already exists, a few general principles can be summarized as follows.

- No single evaluation statistic (e.g. false alarm ratio, probability of detection, root-mean-square error) is capable of presenting a complete picture of the evaluation statistics, depending upon the density and adequacy of the observational networks (Chang and Hanna, 2004). Therefore, multiple statistics should be calculated to develop a better understanding of the model behavior.

- Terminology across the published literature can be used inconsistently (e.g. Barnes et al., 2009), so definitions of the statistics employed should generally be included in any model evaluation study.

- Evaluating higher-resolution forecasts will necessarily result in a relatively worse evaluation relative to lower-resolution forecasts using most of the common statistical parameters (e.g. Roebber et al., 2004). Thus, different approaches need to be considered, especially at convection-permitting resolutions. For more discussion of these particular issues, see the March 2008 special issue on forecast verification in *Meteorological Applications*. In addition, new ways of visualizing forecast quality have been developed (e.g. Roebber 2009).

- Statistical significance of errors should be evaluated and spatial fields should be tested for field significance (e.g. Livezey and Chen, 1983; Elmore et al., 2006a).

Table 10 lists and summarizes some CWF models evaluation activities. A substantial fraction of the evaluation activities included in Table 10 can be classified as diagnostic and dynamic evaluation exercises.

5.3 Multi-model evaluation studies

Model intercomparison studies also offer the chance to see the weaknesses in the models, and thereby lead to efficient improvement. Although all 18 CWF models considered in this article have been evaluated individually by comparison to observations (Table 10), multi-model evaluation projects can tackle some of the problems more cost-effectively. A structured intercomparison among models can also indicate whether a general consensus exists among the models or whether there are outliers.

Multi-model evaluation against field experiments includes the passive-tracer field experiment ETEX (European Tracer EXperiment, http://rem.jrc.ec.europa.eu/etex/), EuroDelta (http://eurodelta.pangaea.de/), CityDelta (Cuvelier et al., 2007, http://aqm.jrc.ec.europa.eu/citydelta/), ESCOMPTE

(Expérience sur Site pour COntraindre les Modèles de Pollution atmosphérique et de Transport d'Emissions; http://escompte.mediasfrance.org), and ESQUIF (Etude et Simulation de la QUalité de l'air en Ile de France, a synthesis of the Air Pollution Over the Paris Region, Vautard et al., 2003). The ETEX experiment is useful for testing meteorological and diffusion processes, but of course not suitable for the evaluation of chemical transformation or deposition of various pollutant species. Multi-model evaluation results are expected to be available from the AQMEII study in near future (Rao et al., 2011).

Three of these projects are described below: EuroDelta, CityDelta and ESCOMPTE. These three model intercomparison projects were selected because they compare several CWF models, including some of those described in this article.

5.3.1 EuroDelta

The EuroDelta experiment was designed to evaluate air-quality improvement over Europe in response to regional emission reduction scenarios for 2020. Within the framework of EuroDelta, van Loon et al. (2007) studied the long-term ozone simulations from seven chemical weather models: CHIMERE, DEHM, Unified EMEP model, LOTOS-EUROS, MATCH, RCG and TM5 (Tracer Model 5); the latter is a global chemistry transport model. The remaining models are regional-scale models using European domain. van Loon et al. (2007) compared the models forecasts to observed ozone concentrations. All modelling groups adopted the same annual emission inventory of ozone, O_x and NO_2 to their model grid and model species.

Most of the models in EuroDelta realistically reproduced the observed ozone diurnal cycle, the daily averages, and the variability in the daily maxima. van Loon et al. (2007) found that "*the daily maxima in ozone concentrations were better simulated than the daily averages, and summertime concentrations were better simulated than wintertime concentrations*". Daytime ozone concentrations were overestimated by all models except for TM5 and DEHM. These two models also had small diurnal cycle. LOTOS-EUROS and RCG, which use the same meteorology and mixing layer concept, had a more-pronounced diurnal cycle than observed. CHIMERE produced a large all-day-long positive bias in ozone concentration, which according to van Loon et al. (2007) may be due to a bias in the boundary conditions. MATCH, DEHM, EMEP and the average of the concentrations from all seven models accurately represented the diurnal cycle of ozone (van Loon et al., 2007).

5.3.2 CityDelta

The aim of the CityDelta project was to evaluate the air-quality response of several emission reduction scenarios in the European continent for 2010, with a focus specifically in the cities (Cuvelier et al., 2007). CityDelta proceeded in two stages. In the first stage, 15 modelling groups participated in the project, with a total of 22 model configurations (e.g. CAMx, CHIMERE, EMEP, LOTOS; Cuvelier et al., 2007). Participants performed a one-year (1999) control scenario simulation for PM and a 6-month simulation for ozone for at leat one of the six European cities selected (Berlin, Katowice, London, Milan, Paris and Prague; Cuvelier et al., 2007).

Within the second stage of CityDelta, Vautard et al. (2007) used the predictions of six models (CAMx, CHIMERE, EMEP, LOTOS, OFIS (Ozone Fine Structure model), and REM-CALGRID) to simulate ozone and PM_{10} concenrations for 1999 around four cities: Berlin, Milan, Paris and Prague. All the models used small-scale grid spacing (5 km), but three of the models (CHIMERE, LOTOS, REM-CALGRID) were used also at large-scale (50 km) grid spacing. A model simulation domain of 300×300 km around the cities were used in the both stages.

The models captured reasonably well the mean, daily maxima and variability of ozone concentrations, as well as the time variability of the ozone response to emission scenarios for each city and the spatial variability between cities. However, the large-scale models overestimated the ozone concentration in the city centres. All models had difficulties in capturing the PM_{10} concentrations and the spatial variability between cities is not reproduced. Especially the large-scale models underestimated the mass of PM_{10} due to the lack of horozontal grid spacing. Vautard et al. (2007) found that the small-scale models show better performance for PM_{10} and ozone concentrations in urban areas than the larger-scale models.

5.3.3 ESCOMPTE

The European campaign ESCOMPTE (Cros et al., 2004) documented four photochemical episodes, lasting 3–4 days each, near Marseilles in the coast of South-East France during June and July of 2001. These days corresponded to about 30 % of the ozone pollution days (120 ppbv or greater) in this region in 2001. The main objectives of the field campaign were to analyze and document several photochemical episodes in this area, as well as to create a detailed chemical and meteorological database for testing and evaluation of regional-scale CTMs. Aerosol measurements were also carried out during ESCOMPTE. The cooperative experimental project was open to all research groups. The objective of ESCOMPTE was not to rank modelling systems according to specific statistical performance, but rather to provide a convenient and comprehensive benchmark to evaluate models or different versions of the models (http://escompte.mediasfrance.org/exercice/HTML/overview.html).

The data from the ESCOMPTE campaign has been used in many studies (e.g. Menut et al., 2005; Coll et al., 2007; Pirovano et al., 2007). Coll et al. (2007) focused on the

simulation of two intense ozone episodes on 21–23 June 2001 and 24–26 June 2001 using two models, CAMx and CHIMERE, with several configurations. The results of all the model configurations were examined to determine how much the changes in dynamical and chemical input data affected the models outputs, in an attempt to discriminate between the influence of internal and external configuration choices. One conclusion of Coll et al. (2007) was that ozone plumes are strongly influenced by the modelled representation of the wind circulation, because the structure of the ozone plume over the domain was driven by wind fields.

6 User operations

This section provides an overview of how the user interacts with the different models that produce the operational forecasts. Section 6.1 discusses the availability, documentation and user interfaces of the different models. This section also discusses the computer requirements and lists the levels of documentation. Section 6.2 discusses how the output is disseminated. Section 6.3 describes internet portals where CWF model output is disseminated and presented. A summary of the availability, user communities, and documentation of the various CWF systems is presented in Table 11.

6.1 Model availability and documentation

The availability of CWF models, and more specifically their source code and documentation, may be described in terms of software availability, and their use may be categorized in a similar way. On this basis, many of the models are provided as free and open-source environmental software (Karatzas and Masouras, 2004), such as CAMx, CHIMERE, CMAQ, MM5, SILAM and WRF-Chem. In contrast, other models are not publicly available or are otherwise restricted in some way, such as Eta, MEMO, Unified Model, GME, ALADIN, EURAD, FARM, MATCH, MOCAGE, NAME, OPANA, SKIRON/Dust and THOR. There are also models that combine public and restricted source codes, such as CAMx-AMWFG, Enviro-HIRLAM and MM5-CHIMERE.

The terms of use for those models that are not freely available are not identical for all categories of users; research institutes are usually not charged for their use, although this may not be the case for commercial applications. In addition, model availability options may also exist, as in the case of free access being limited to institutes participating in a specific consortium, or in the case of a distinction between the operational version and the research version. Some models may have well-organized and regularly updated web sites, including documentation on model applications, evaluation and user communities. In contrast, others suffer from poor or incomplete documentation.

The level of documentation of each model included in Table 11 is ranked according to a five-level scale based on the relevant information provided by the Model Documentation System (MDS) of the European Environment Agency (http://pandora.meng.auth.gr/mds/help/lh_documentation.html). The MDS is an on-line database for CWF models that provides various search facilities and a structured, homogenized display of model information. This system has been available for the last ten years (http://pandora.meng.auth.gr/mds/mds.php, Moussiopoulos et al., 2000). Another internet-based system of model properties is in the joint COST 728 and COST 732 Model Inventory, accessible at http://www.cost728.org (Schluenzen and Sokhi, 2008).

Because CWF models are computationally intensive, they are not usually prepared as a software product ready to be installed and executed. Sometimes various software tools may be used that commonly accompany the source code and are usually described in the model documentation. Few model packages have dedicated user interfaces that allow for the automatic installation, set-up and use of the model. Commonly, command-line scripts (usually shell scripts) and compilers are required to produce an operational executable. In addition, using CWF models requires software tools for the preprocessing (e.g. input data preparation, formatting, autofeeding), as well as post-processing (e.g. model visualization) of model output data.

6.2 Users of CWF model results and information dissemination

CWF models require a high degree of expertise to be used, applied and configured, and interpretation of their output also requires some experience. Due to the complexity of CWF models, the need for the training and support of CW users is absolutely essential. Examples of training courses and programs can be found in the past for models such as CMAQ, and general courses for training personnel in different institutes and commercial companies have been developed in the past from American and European universities and research laboratories.

CWF model users are usually scientists who set up and execute the model for operational or research uses and produce model results. However, CWF model results are of interest for many other categories of users. Specifically, the importance of this output may be traced to the environmental regulatory and legal framework, the resulting mandate for improved air-quality management, and the interest of citizens in environmental issues.

In Europe, CWF has been guided by a number of directives – which have been implemented by different agencies or government bodies in different European countries – that define the quantitative thresholds to be applied to address air-pollution problems. The latest update of this legal framework is related to the adoption of the Clean Air for Europe Directive 2008/50/EC, which states that "*Member States shall ensure that timely information about actual*

Table 11. A summary of the availability, user communities, and documentation of the various CWF systems.

Model name	User interface availability	User community	Documentation status	Availability
ALADIN-CAMx	Mainly Linux shell scripts and Fortran90 (gfortran) code	Researchers for regulatory applications. Combination of ALADIN-Austria and CAMx is only implemented at ZAMG	Level 1: CAMx (www.camx.com), physical description of model, code administration, installation guide, evaluation. Level 1: ALADIN (www.cnrm.meteo.fr/aladin), physical description of the model, variable settings	Not a public domain programme. Information on the conditions for obtaining the model can be provided by the contact person. CAMx is open source software: www.camx.com
CAMx-AMWFG	Linux shell scripts, Intel Fortran using OpenMP. NCAR graphics	The model can be used properly by a highly skilled person	Level 1: complete documentations available, ranging from the scientific description down to users manuals with details on the machine code	CAMx-AMWFG (with the new features) availability should be discussed with the contact person. CAMx is open source code available from www.camx.com
Enviro-HIRLAM	Shell scripts and FORTRAN code. METGRAF graphics	Enviro-HIRLAM is baseline system for the international HIRLAM chemical branch. The model can be used properly by a highly skilled person	Model formulation, practical lectures notes and web-description. Level 2, English. www.hirlam.org/chemical	For users of the HIRLAM chemical branch. Enviro-HIRLAM is an open community model, source code is available from hirlam.org upon agreement
EURAD-RIU	Linux shell scripts, Portland Fortran compiler using OpenMP or MPI, NCAR Graphics	The EURAD research group at RIU	Model description and user manual	Not a public domain model system. Availability possible within common projects
FARM	Post-processor for statistical analysis of modelled fields and data extraction as well as interface with visualization tools (AVISU, Savi3-D, GrADS, Vis5D)	Italian national and local environmental authorities, research institutes. Users of FARM should have a sufficient background in atmospheric sciences and experience in the use of complex numerical models	Model formulation and user manual. Level 1-2, English	Available to selected users
LOTOS-EUROS	Configuration file including description, shell scripts to compile and start	Scientists within national and international projects. Forecasts provided to the public	Yearly reports with description of physics and parameterisation. Technical documentation for daily usage	Limitted to institutes participating in model consortium. Information on the conditions for obtaining the model can be provided by the contact person
MATCH	Shell scripts and Fortran code, requiring technical expertise. More user-friendly frontends with less configuration options exist for specific applications	Scientists within national and international projects. Environmental agencies and decision makers (access mostly through tailored interfaces). Forecasts provided to the public	Level 2: rather good scientific documentation and less complete user's manuals. Automatically generated html-documentation included in model installation. General information also available on the web-page	The model is not a public domain programme. Information on the conditions for obtaining the model can be provided by the contact person
MM5-CAMx	Linux shell scripts, Intel Fortran using OpenMP, PAVE and GRADS visualization tools	Research groups, environmental agencies, private companies. The modelling system can be applied by skilled users	Complete documentation (scientific description, user's guide, software) available at: MM5: http://www.mmm.ucar.edu/mm5; and CAMx: http://www.camx.com	The MM5-CAMx system used for air-quality forecast in Europe, in the Balkans and in Athens is not publicly available. The availability of the system can be discussed with the contact persons. Both MM5 and CAMx are publicly available models
MM5-CHIMERE	CHIMERE: Fortran program called CHIM2FIG for automatic generation of maps, vertical cross-section, time-series using GMT free software. The exachim tool using Grads free software	CHIMERE users mailing list: chimere-users@lmd.polytechnique.fr Website: http://euler.lmd.polytechnique.fr/chimere Questions: chimere@lmd.polytechnique.fr	MM5: journal publications, workshop preprints, NCAR technical notes and manuals are available from the NCAR MM5 Community Model Homepage. CHIMERE: complete documentations available, ranging from the scientific description down to users manuals	MM5: the model is a public domain programme. The source code and utility programmes can be downloaded from the NCAR MM5 Community Model Homepage: http://www.mmm.ucar.edu/mm5/mm5v3/wherev3.html CHIMERE: online free access to the code under the General Public License
MM5/WRF-CMAQ	Shell and Tcl shell scripts. IDV and PAVE visualization tools. Interfaces developed to convert output original files (NETCDF) to Ferret format	Research groups, environmental agencies, private companies. The modelling system can be applied by skilled users. Operational air quality forecasting systems for cities and industrial plants. Used for air-quality impact studies for new power plants, incinerators and oil companies	All documentation can be found in http://www.cmascenter.org/. MM5 was originally developed by NCAR/PSU in US and the development of WRF is a collaborative partnership among the NCAR, NCEP, FSL, AFWA, the Naval Research Laboratory, University of Oklohoma, and FAA. CMAQ was originally developed by US EPA	MM5/WRF-CMAQ is in the public domain. Scripts developed ad-hoc for running the systems and for specific customized applications have to be developed on demand. Please contact Director ESMG or CAIR, University of Hertfordshire for details

Table 11. Continued.

Model name	User interface availability	User community	Documentation status	Availability
MOCAGE	UNIX/Linux shells or PrepIFS/ Olive interfaces to run the model. Output is available in NetCDF (CF) or/and LFI or GRIB. Tools (fortran code, shell script in UNIX/LINUX or PYTHON) are available for graphical production with GMT, FERRET and IDL	Météo-France research centre, Météo-France operational departments (environment and health division, forecasters in support of accidentel crises), research labs (Laboratoire d'Aérologie, CERFACS), PREVAIR users (air-quality agency, ministery for environment,...), National Met Services (AEMET Spain, DMN Morroco, NMA Roumania, KMA Korea)	In addition to peer-reviewed international litterature, model documentation (scientific, user) is currently composed of several documents. Harmonization of these and further developments are on-going. Detailed information on demand	The code is not in the public domain, but it is available free of charge for research purposes under a specific scientific collaboration agreement. For operational and/or commercial application of the code or outputs, a negociation is needed with Météo-France (contact us)
NAME	Simple text file	UK Met Office Universities, UK government and defence organisations	Level 2/Level 3 documetation. Adequate user manual. Significant evaluation of model against experimental data	The model can be made available to research and commercial users via negotiation with the Met Office
OPANA	Friendly user interface which can be easily customised to user specifications	City Council, Environmental Administrations or Regional Authorities, industry, etc	Model is having an on-line help and on-line manual	Conditions to have a licence of OPANA framework can be provided by the contact person
RCG	Input ascii-file must be compiled	Governmental or local authorities, scientists within national and international projects	Reports and publications. Documentation available on demand	No info
SILAM	Available for emergency tasks (restricted access via SILAM Web portal)	Operational dept. of Finnish Met. Inst. (Emergency preparedness, Operational air quality forecasts), Finnish Radiation Protection Authority (Emergency preparedness). Research purposes: Finnish Met. Inst., and other Research groups	Level 1 documentation for the operational version is available from the Web site (http://silam.fmi.fi/)	The model is public. Well-tested operational version v.3.6.5 is available from the Web site. Research version of the model is available on request
SKIRON/Dust	Linux shell scripts, Intel Fortran using MPI parallel processing. NCAR graphics	The model can be used properly by a highly skilled person	Level 2: rather good scientific documentation and less complete user's manuals. General information also available on the web-page	Information on the availability can be provided by the contact person
THOR	Linux shells, Pathscale, portland and Intel Fortran, UNIRAS graphics, vis5d, IDV, NCL	Research groups. The modelling system can be applied by skilled users	Peer review papers and some documentations available	Not a public domain model system. Availability possible within common projects
WRF-Chem	Shell and Tcl shell scripts. IDV and PAVE visualization tools. Interfaces developed to convert output original files (NETCDF) to Ferret format	WRF-Chem is used as a research tool by ESMG-UPM. Recently applied for internal tests in operational air-quality forecasting systems. Applied in several COST 728 projects	Documentation available at http://www.mmm.ucar.edu/wrf/users/supports/tutorial.html	WRF-Chem is in the public domain. Scripts developed ad-hoc for running the systems and for specific customized applications have to be developed on demand. Please contact Director ESMG for details

or predicted exceedance of alert thresholds, and any information threshold is provided to the public". On this basis, a set of CWF goals is defined that include the reporting of the geographical area of expected excedence of an air-quality threshold, the expected changes in pollution (such as improvement, stabilization or deterioration), and the reasons for those changes.

Moreover, the same directive states that "it is necessary to adapt procedures for data provision, assessment and reporting of air quality to enable electronic means and the Internet to be used as the main tools to make information available". This means that it is necessary to develop *operational* air-quality management and citizen notification systems that will make use of modern information and communication technologies and will allow for the *early forecasting* of air pollution levels (Karatzas, 2010). This means that environmental authorities are required to operate systems that will include operational CWF models, and would allow them to estimate the spatial and temporal occurrence of air pollution, in advance of any actual incidents, and thus notify citizens and other interested parties.

Directive 2008/50/EC establishes the need to measure the uncertainty for a CWF model applied to a specific area, as well as the uncertainty of monitoring stations. It is important to take into account that scientific community, policy makers and citizens have relative different approaches to categorize and interpret model skill and the use of different categorical statistics such as hits, misses and false alarms (e.g. Barnes et al., 2007, 2009).

The users of CWF model output may be defined with the aid of the air-quality information provision requirements of the 2008/50 EC Directive, as well as from common practice (e.g. Fedra and Witner, 2009; Slørdal et al., 2008; Karatzas and Nikolaou, 2009). Specifically, we provide some examples of how CWF models are currently being applied by end users.

6.2.1 Industry and business end-users

The main interest of these users is to forecast the impact of industrial emissions from installations such as power plants and petroleum refineries. As these users are interested primarily in the output of CWF models, it is easier for them to commission these forecasts as a service rather than having to install and maintain the modelling system themselves.

One important category of the use of CWF model results is the assessment of air quality in industrial areas and their surroundings. The spatial scale in such applications may be of the order of from tens to hundreds of kilometers, and the temporal scale of the order of days, or years in case of forecasts for future scenarios. For example, in the case of a multi-source industrial complex, the MM5-CMAQ air-quality management system may be applied (San José et al., 2006, 2008a, b). Due to the computational demand of such problems, a computer cluster or a multi-processor machine are among the most appropriate hardware set-ups, where the models are run in parallel, handling different emission scenarios. For these applications, the service provider usually prepares the software for managing the simulations, as well as the software required for the pre-processing of the input data and the post-processing of the results and the necessary web-based interfaces for the client. Such systems provide decision-making support for clients, who need to decide whether to switch off some emission sources, usually within the next 24 h, to avoid an air-quality episode.

6.2.2 The environmental decision and policy makers

These users are responsible for making decisions about air-quality abatement measures, as well as managing air-quality status and dealing with problems on a local to regional scale. An important category of users is city authorities. Clearly, city authorities are interested in the capability of the modelling system to forecast all the parameters that are required by the relevant regulatory framework, as well as the accuracy and accountability of the information that is being produced. In both cases, information dissemination is usually based on the automatic (or semi-automatic) preparation of tables and graphs, providing estimates of concentrations, their spatial and temporal evolution, and scenario-based estimates of the emissions and meteorology.

Designing and predicting forecast-based scenarios is important for decision making, as it allows authorities to take preventive measures to avoid an air-quality episode or reduce the duration or spatial scale of a forecasted episode, in accordance to the mandates of the 2008/50/EC Directive. Originally, many city authorities had maintained and operated their own operational CWF modelling systems. However, due to the increased complexity of the latest versions of such systems and the capacity required in terms of experienced personnel and hardware, the tendency now is to hire such services from partners such as institutes, universities or private companies that are active in this area, or to install the system locally and contract with consultants for services, upgrades, and maintenance.

6.2.3 The CWF scientific community

The CWF scientific community has a strong interest in the science and the understanding of CWF phenomena and problems. This community requires detailed information, which is usually of little or no interest to the other user communities. This information includes model performance indicators, model improvements, and environmental decision-making analysis data. Nevertheless, the detailed results of the operational CWF model calculations are generally not made available to those outside the group that has developed and is maintaining the CWF modelling system.

6.2.4 The general public and susceptible populations

These include inhabitants of the area covered by the operational CWF models, as well as people living outside this area, who nevertheless are interested in the air-pollution levels near the area where they live or work. The susceptible populations comprise of children, the elderly, and adults with respiratory, cardiovascular or other relevant impairments. For these users, CWF models are combined with air-quality information systems, that make use of complementary push-pull communication channels (Zhu et al., 2002; Karatzas et al., 2005; Karatzas, 2007; Karatzas and Nikolaou, 2009).

The dissemination of the air-quality information to the general public is usually in the form of air-quality indexes, graphical representations of air-pollution levels, text descriptions, and multimedia. The means of dissemination are quite variable, including mass media, Internet, personalized SMS (short message system) messages, voice servers, and dynamic street-level displays. These characteristics have also resulted from the analysis of a set of air-quality information dissemination systems that was conducted under COST Action ES0602 (www.chemicalweather.eu; Kukkonen et al., 2009a). This COST Action inventoried the way that air-quality information was disseminated to the public by analyzing data from 93 air-quality information systems, originating from seven European countries (Karatzas and Kukkonen, 2009). The air-quality information systems that were screened were divided into two types:

1. Those that disseminate air-quality information based on observational data. In many of the systems analyzed, air-quality observations are provided to the public on the basis of hourly data. In some cases, this information is made available in near real time (with a time lag of 1–2 h), whereas, in other cases, this information is provided for the previous day, or up to the last period, for which data have been evaluated.

2. Those that disseminate air-quality information based on operational CWF model forecasts. However, in many of the air-quality information systems investigated, no CWF models were applied. This suggests that the CWF modelling community needs to apply models, not only for regulatory purposes, but also for producing information for all three categories of users. In the cases where CWF models were applied, these were mostly three-dimensional models, although statistical models and computational-intelligence models were also employed in some cases instead of three-dimensional models. In some cases, human judgment is applied to estimate the quality of the atmospheric environment for those systems that have no CWF model support, whereas in some cases both human expertise and models are used.

6.3 Dissemination of forecasts on the Internet

To investigate the basic characteristics of operational CWF modelling systems, an analysis was made based on the systems that are currently included in the European Open Access Chemical Weather Forecasting Portal, which has been implemented within the framework of COST Action ES0602 (Balk et al., 2011; available at http://www.chemicalweather.eu/Domains, and registered as a GEOSS service in 2010). This portal provides access to available CWF systems in Europe in a user-friendly graphical format. The portal currently includes about 20 operational CWF modelling systems (12 included in this article) from across Europe, covering local to regional and continental scales of air quality. The basic characteristics of these systems are summarized in Table 12.

In all these studied systems, users only had to make one or two selections to obtain the information (in terms of graphs or maps). Many systems do not archive forecasts, whereas others archived information for the last two days or two months, and others archived years of data.

The Internet is the most popular way to disseminate output from operational CWF models. For the models in the European Open Access CWF Portal, many provide output in the form of concentration fields, usually superimposed on maps of the area of interest. The use of coverages (i.e. two-dimensional pollution images) for the presentation of CWF results is the most popular method of relevant information dissemination. Such information usually includes various pollutants and refers to the surface (ground) layer, as well as in some higher vertical levels. Such output graphics are always geo-referenced (covers a specific geographic area) and time-stamped (usually of to 72 h ahead, in hourly intervals). Commonly, multiple CWF models produce information for the same geographical area and time. Yet, as every model uses its own color-scale for mapping pollution levels, geographic projection and other parameters of information presentation, it is not easy for the end user to compare information coming from different CWF systems. Importantly, model output is usually not accompanied by any quality and reliability indicators related to the forecasting performance of the model and the robustness and trustworthy of the forecasting service.

The seamless and comparable access to CWF results, quality of information, and reliability of service are considered to be the most important factors for the use of forecasts by every user community. In addition, different user communities have different interests in forecasting products. Citizens and the general population are usually interested in exceedances, their intensity, duration and location, as they would like to safeguard as much as possible their quality of life. For the same reason, users also prefer simple, intuitive methods of information presentation (e.g. graphs, color scales), and they do not like to receive graphs, tables and any type of information that requires additional expertise to be interpreted. They also prefer CWF products that are tailored to everyday

Table 12. Dissemination of CWF system predictions in the Internet. The results correspond to characteristic properties for European regional-scale CWF systems (Balk et al., 2011).

Operational CWF modelling system characteristic	Parameters and explanation
Forecasting period	The time frame of the forecasting, ranging from 24 h to 3 days in advance
Pollutants forecasted	Two to seven pollutants were addressed, and include $PM_{2.5}$, PM_{10}, NO, NO_2, NH_3, O_3, SO_2, CO, benzene, and radon
Information type provided	Varies on a case-by-case basis, and includes daily mean, daily maxima, hourly values, hourly averages, hourly maxima, 8 h running average (for Ozone), and Air-Quality Index
Information presentation	In the majority of cases, the information is presented with the aid of two-dimensional pseudo-color concentration contours. Some times are available as animations or spot maps. Images are GIF formatted, and in a few cases are available also as PNG files, or via a Java Applet
Additional information	Some systems also provided information on the road and railroad network, wind speed, cloud coverage, temperature, mixing layer, animated trajectories, wet deposition, and time series graphs for selected locations. Animations of various days were also available for some parameters ion some systems
Web site technology and user interface	In the majority of cases, this was covered with HTML and AJAX. Although in many cases, the solution adopted was HTML and PHP, or the applications used HTML and Java, HTML, AJAX and Java, HTML, or PHP and AJAX

human activities, such as commuting and recreation, as was found for mesoscale weather information in the Helsinki Testbed (Koskinen et al., 2010). Policy makers prefer products that may help them in environmental management and decision-making tasks such as the number of exceedences and the most affected areas. Another useful product is a scenario-based analysis of alternatives that may help identify the potential of preventive or abatement measures (so-called it what-if scenarios). Scientists and CW experts prefer products that help them in developing a better understanding of the underlying phenomena, in analyzing the mechanisms (physical, chemical, etc.) employed in CWF, and in evaluating the accuracy of models and model ensembles.

A common demand by each category of users is that model results are made available seamlessly and in advance and that they be accompanied by near-real-time data coming from actual measurements. Overall, there is a growing demand for a service-oriented approach in CWF that can tailor related products to user categories in a more effective and reliable way.

7 Emerging areas and future challenges

The aim of this section is to highlight selected emerging scientific areas, as well as future challenges that would be expected to lead to improving the reliability of chemical weather forecasts. These topics include emission and chemistry uncertainties (Sect. 7.1), integration of NWP and atmospheric chemistry transport models (Sect. 7.2), boundary conditions (Sect. 7.3), assimilating chemical data into the models (Sect. 7.4), improved understanding and parameterization of physical processes (Sect. 7.5), evaluation of CWF models against data (Sect. 7.6) and generation of model ensembles (Sect. 7.7).

7.1 Emission inventories and modelling

The evaluation of emissions is one of the main sources of the uncertainties in the predictions of the CWF models. In this section, we address the research challenges in terms of the pollutants and source categories, and in terms of how various emission inventories should be refined and harmonised.

7.1.1 Research challenges of emission inventories of species and source categories

Improvement is required for the emission inventories, particularly of PM and organic species. Most of the regional emission inventories currently consider PM_{10} and $PM_{2.5}$; however, primary aerosol emissions need to be further specified in terms of the aerosol size distributions, chemical composition and source origins. Clearly, all the chemical constituents of PM are needed for particle mass closure, for determining the physical and chemical properties of PM, and for comparison with speciated PM concentration and precipitation-chemistry measurements. In

Table 13. Brief demonstration of some sensitivity analysis applications with the examined CWF models.

Model	Uncertain inputs	Method	Main findings	References
CAMx	Initial and boundary conditions, emissions	Direct Decoupled Method, Tangent Linear Model	Ozone changes predicted with the DDM sensitivities were compared to actual changes obtained from simulations with perturbed by 40 % inputs. The DDM sensitivities converged toward the brute-force sensitivities for the perturbations in initial or boundary concentrations while for the perturbations in VOC and/or NO_x emissions, the magnitude of the predicted changes is 10–20 % smaller than the actual changes on average	Dunker et al. (2002)
CAMx	Boundary conditions, area and point emissions	Tangent Linear Model	The importance of the studied uncertain inputs varied among the examined metropolitan areas (Athens, Milan, London) during the selected episode days corresponding to the 98th annual ozone percentile. However, the variation in only two inputs, namely the boundary conditions of O_3 and the area emissions of NO_x was found to produce the highest change in ozone concentrations in the examined urban areas. In terms of validation, the linear perturbation fields tangent linear model represented very well in both location and magnitude the expected values for at least 25 % reduction in NO_x and VOC emissions and for at least 50 % variation in the boundary conditions of ozone.	Kioutsioukis et al. (2005)
CHIMERE	Boundary conditions, emissions, reaction rates, meteorological fields, dry deposition	Adjoint Model	The results point out three types of model parameters to which the concentrations are very sensitive: (i) the boundary conditions (mainly ozone, PAN and HCHO seem to have a relevant effect on the simulated concentrations), (ii) the meteorological fields (such as temperature, wind speed and vertical diffusivity) and (iii) the surface emissions (NO_x in the urban areas and VOC in the rural areas). On the other hand, dry deposition was found to be not so sensitive.	Menut (2003)
CHIMERE	Boundary conditions, urban emissions and chemical reaction rates	Adjoint Model	The ozone peak, for this particular day, is essentially sensitive to traffic and solvent emissions (and about in the same ratio). The sensitivity to reaction rates is fairly nhomogeneous: only a few reactions are sensitive, among which the photochemical equilibrium between NO_x and ozone, the reaction of NO_2 with OH, the photolysis of ozone and aldehydes and the oxidation of reactive primary hydrocarbons. In terms of ozone boundary conditions, it was found that a correct estimation of these fluxes is of crucial importance for a correct simulation of the ozone concentration in the urban area (Paris).	Vautard et al. (2000)
CMAQ	Emissions, temperature	Adjoint Model	The efficiency of the CMAQ adjoint was demonstrated in this study through several examples. In particular, adjoint analysis can for example: (i) identify the most influential emission sources that contribute to the overall population exposure, (ii) quantify the impact of increased ozone on crops and vegetation and in addition indicate the sources where emission control can result in largest reductions in the environmental exposure metric, (iii) measure the effect of temperature variation on air pollution levels to e.g. formally quantify the impact of future climate conditions on regional air quality.	Hakami et al. (2007)
EURAD-RIU	Emission rates	Adjoint model (4D-var)	The objective of the present study was to test the potential of the 4D-var method for emission rate optimization. It is demonstrated that in the case of NO the emission rates can be estimated, provided the first guess is not too far from the locally governing chemical regime. Emission rates of VOCs could not be analyzed individually. However, by adopting regularization techniques, well established a priori knowledge of exhaust VOC composition is introduced and a skillful analysis could be obtained. In addition, temporal variations of the emitting sources could be retrieved. In general, it can be concluded that four-dimensional variational parameter estimation appears to be a promising tool to analyze emission rates of various emitted, but not observed, species, if some reasonable regularization assumptions can be made.	Elbern et al. (2000)

Table 13. Continued.

Model	Uncertain inputs	Method	Main findings	References
MOCAGE	Deposition velocities	Brute force	The highest maximum relative differences are found for HNO_3 (17 % mean), with the largest differences to appear in the troposphere. The next species in terms of large relative differences is NO_2 (10 % mean). Then comes OH and O_3 with means of 5 and 2 % respectively. For O_3, the relative differences decrease rapidly from the surface up to about 800 hPa; the highest differences appear in May. Finally, all maximum relative differences are below 6 % for CO throughout the whole atmosphere. As for ClO and HCl they have non significant relative differences in the troposphere due to their very small mixing ratios, and relative differences lower than 10 % in the stratosphere. For NO_2, relative differences are almost nil throughout the atmosphere. The variability of the HNO_3 deposition velocity is high as it is driven by the aerodynamic resistance and thus the stability of the atmosphere.	Teyssèdre et al. (2007)
SILAM	Meteorological input data, model setup	Adjoint model	The study allowed selection of the optimum setup for the operational model configuration. The most important factors with regard to the model performance were (i) the selection of the meteorological input dataset and (ii) the method used for the atmospheric boundary layer height estimation. Specifically, the sensitivity runs with different sources of input data showed that high spatial and temporal resolutions do not automatically lead to better results. Further, it was found that use of the PBL height estimate from the NWP is the best option.	Sofiev et al. (2006a)

particular, particulate black carbon, organic carbon, and organic mass should be specified. Clearly, all aerosol chemical components, originated from various source categories, should be included in order to obtain an aerosol mass closure. Reff et al. (2009) have provided a review of the speciated PM emissions. Natural emissions of PM – for example, duststorms in arid or semi-arid areas, wild-land fires (e.g. Saarikoski et al., 2007; Sofiev et al., 2009; Saarnio et al., 2010) and sea-spray – are emerging areas of further research. In particular, information is scarce on the size distribution of particulate matter formed from natural dust sources and the temporal variability of dust emissions.

Substantial progress has recently been achieved in the representation of processes controlling biogenic VOC emissions (Monks et al., 2009). However, biogenic VOC emission models still need improved quantification by species type (e.g. isoprene) and increased number of species in inventories. Also numerous semi-volatile and low-reactivity organic compounds that contribute to secondary organic aerosol formation are not commonly included in the inventories. Information on the emissions of residential and other small-scale combustion is scarce, although its influence on the exposure of the population may be substantial in some countries and regions (e.g. Karvosenoja et al., 2008, 2010; Denby et al., 2010).

Major uncertainties remain for emissions from transport, including emissions from shipping and aviation, and on the vehicular non-exhaust emissions. For example, not all emission inventories consider ship emissions, which can be important to air pollution in coastal areas (e.g. Jalkanen et al., 2009 and 2011). Also, uncertainties remain in the modelling of emissions that are dependent upon meteorology, such as allergenic pollen (e.g. Sofiev et al., 2006b, 2011a; Veriankaitë et al., 2010) and dust.

The man-made or natural activities that give rise to discharges of various substances into the atmosphere are identified in a reference list known as SNAP (Selected Nomenclature for Air Pollution). SNAP is structured on three levels: source sector, sub-sector and activity. Currently, the existing gridded anthropogenic emission inventories over Europe identify SNAP level 1 emitting sources. Improvement is required in the availability of European spatially resolved anthropogenic emission data for SNAP level 2 (and 3) emitting sources. Also, detailed libraries of temporal profiles and speciation profiles associated with these emitting sources could allow better temporal allocation and chemical speciation of pollutant emissions to be used in CTMs applications.

7.1.2 Research challenges on harmonisation of emission inventories

The lack of harmonisation of emission inventories at European and national levels is one of the main obstacles to the quantitative comparison of the predictions of operational CWF systems. Currently, the horizontal grid spacing of the emission inventories can be reasonably accurate for regional CWF systems (e.g. the grid spacing for the pan-European

domain is 7×7 km in the emission inventory within the MEGAPOLI project). However, the temporal variability of emissions and the vertical distribution of the heights of the emission sources are not considered accurate in all cases, and these aspects of the emission inventories need to be improved.

Further work is also needed to improve the relationships between global, regional and local inventories, especially for developing countries and urban areas. Global emission inventories (e.g. EDGAR, Emission Database for Global Atmospheric Research; IPCC/IIASA and Intergovernmental Panel on Climate Change/International Institute of Applied System Analysis) result in major uncertainties for the total emissions of individual major cities. For example, non-methane hydrocarbon emissions for London as specified by the various available inventories differ by about 65 %; for Moscow and Paris, they differ by almost a factor of three (Gurjar et al., 2008). Even for NO_x emissions, the emissions for Paris differ about by a factor of 2.5 and for Moscow more than 60 % (Gurjar et al., 2008).

The global emission inventories commonly underestimate the urban emissions relative to national and municipality databases, as is the case, e.g. for the London Atmospheric Emission Inventory database (LAEI, 2009). A quantitative analysis of such differences is therefore needed, and more accurate emission inventories are required on regional and city levels. The first step in this direction was taken within the European CityDelta project (Sect. 5.3.2; Cuvelier et al., 2007). Within the MEGAPOLI project, a new emission inventory has been developed for Europe and the world, with downscaling to urban areas at a horizontal grid spacing of down to 1 km (van der Gon et al., 2009).

When CWF models are used with European gridded emission inventories on a smaller domain, numerical errors are caused due to grid interpolation (as there may be different grid spacings and orientation, and map projections). Clearly, such errors can result, even if the CWF model applies a grid spacing that is similar in magnitude, compared with the larger domain grid spacing.

7.1.3 Research challenges on the temporal variation and satellite observations in emission modelling

Because CWF models typically use emission inputs with data every hour, emission models are used for characterization of daily, weekly, monthly and yearly cycles of sources or their categories. For anthropogenic sources, these models are usually static and simple. Typically, multiplicative coefficients are used to calculate proportions of the total annual emissions appropriate for a given month, weekday and hour. The next generation of dynamic anthropogenic emission models could take inspiration from energy consumption models, which take into account meteorological variables, especially ambient temperature, cloudiness and wind speed. For example, combustion, which is one of the key emission sectors, could be represented in this manner. For operational air quality forecasting, the available inventories are always retrospective and never current. Similarly, in non-operational research projects, the availability of sufficiently up-to-date emission inventories is commonly a challenge, for instance, in the simulations of project field experiments.

Satellite instruments (e.g. OMI, GOME-2 – the Global Ozone Monitoring Experiment, MODIS – Moderate Resolution Imaging Spectroradiometer, MOPITT – Measurements Of Pollution In The Troposphere) provide new opportunities for evaluation and data-driven estimates of emissions. Burrows and Borrell (2009) provide an overview of different instruments. Standard approaches to analyzing satellite data often involve comparing long-term averages of satellite-retrieved columns with simulated columns based on a CWF model. This approach can also be used to evaluate emission inventories. Another approach to estimate the long-term trends in emissions is the so-called analog approach where trends in observed columns are compared to trends based on inventories (e.g. Konovalov et al., 2008). Adjoint (i.e. inverse) dispersion modelling can also be used to evaluate the emissions or the sensitivities of concentrations with respect to changes in emissions (e.g. Tanimoto et al., 2008; Kurokawa et al., 2009), or even to reveal inaccuracies or mistakes included in emission datasets (Prank et al., 2010).

7.2 Improved integration of NWP and atmospheric chemistry transport models

Historically, air-pollution forecasting and NWP were developed separately and the corresponding communities had limited contact and cooperation. Although this situation could be tolerated in previous decades when NWP data were rarely available operationally for chemical weather forecast models and the resolution of NWP models was too coarse for mesoscale air-pollution forecasting, this situation has changed during this century as modern NWP models approach or include mesoscale and city-scale resolution. This progress has been possible due to advances in computing power, high-speed computing networks and the availability of land-use databases and remote-sensing data on a finer resolution.

As a result, the conventional concepts of air-pollution forecasting may need revision, as greater integration is required between NWP models and atmospheric chemical transport models. Several national meteorological services (e.g. Environment Canada, DMI, FMI) have suggested extending meteorological weather forecasting to environmental forecasting that includes both NWP and CWF. Clearly, this concept would ideally also include biological forecasting, such as allergenic pollen species (Kukkonen et al., 2009a, b, c; Baklanov et al., 2010b).

The on-line integration of NWP or other meteorological models with atmospheric chemical transport and aerosol models has several advantages. Such an integration provides

the opportunity to use all three-dimensional meteorological fields in CTMs at each time step and to include feedbacks of air pollution (especially those due to aerosol particles) onto meteorological processes. Extensions into climate modelling include the feedbacks between air pollution and climate forcing, as well as the atmospheric chemical composition. Such a future research direction could be viewed as part of a step towards Earth Modelling Systems and could potentially lead to a new generation of models for NWP and CWF (Baklanov, 2010b).

However, the on-line approach is not the best way for model integration in all cases. For some tasks, such as for emergency preparedness, when NWP data are available, off-line coupling can provide results more quickly. Both off-line and on-line coupling of NWP models and CTMs are therefore useful. A future research area will therefore be to assess the interfaces of these two categories and to establish a basis for their harmonization and benchmarking.

The communication between off-line coupled meteorological and chemical weather models is a problem of often-underestimated importance. The multitude of modelling systems previously introduced gives rise to different approaches and methods implemented within interface modules. Tasks covered by interfaces are minimized in coupled systems. Other systems use interface modules that implement surface and boundary-layer parameterisations to estimate dispersion parameters. Sometimes these latter choices are due to the need to rely on conventionally used meteorological products and to guarantee the robustness of chemical weather modelling for practical applications.

In other cases, interfaces are used to enhance the resolution of local physiographic data and possibly to introduce advanced parameterisations (e.g. those for the urbanisation of models). Atmospheric physics parameterisations – and even default and upper- or lower-limit values assumed for some key parameters – can have effects on pollutant concentration fields in critical conditions (e.g. low wind-speed conditions, stable conditions). Moreover, interface modules may involve the evaluation of emissions of species that can be substantially influenced by meteorology, such as biogenic VOC, windblown dust and sea-salt spray.

Improvements in CWF will also come from assimilating physical parameters that will lead to better estimates of clouds and mixing-layer heights. For example, the assimilation of satellite-derived skin temperatures can be used to better determine heat capacity and moisture fraction of grids. Such data can fill gaps in diurnal energy budgets, resulting in improved short-term forecasts of temperatures, mixing heights, clouds, and photolysis rates (McNider et al., 2005; Arastoo et al., 2007). The recently established new COST Action ES1004: European Framework for On-Line Integrated Air-Quality and Meteorology Modelling (EuMetChem) will focus on further development of integrated CWF systems and on the new generation of on-line integrated chemistry and meteorology models with two-way interactions between atmospheric chemistry (including gases and aerosols), clouds, radiation, boundary layer and other meteorological and climate processes.

7.3 Boundary conditions and nesting of CWF models

An important aspect in the regional applications of CWF models is the type of initial and chemical boundary conditions used by CWF models. The use of climatological averages is one of the common practices, but implementing boundary conditions obtained from global chemical weather models is currently a significant challenge (Tang et al., 2007). This challenge consists of obtaining the required parameters (especially regarding the properties of particulate matter) from the global model within enough time and at sufficient resolution to produce a real-time forecast. Another emerging research area is the development of optimal nesting techniques of CWF models from the global to city scale, using one- or two-way nesting techniques, with boundary conditions on the inner domains provided from larger-scale model domains. For instance, in the PASODOBLE project, most participating models are first used to compute European-scale air quality using global chemical boundary conditions from the global MOZART model from the MACC project, and then applied using nesting inside the same participating model.

Chemical boundary conditions from global CWF systems are already provided operationally to some regional CWFSs around the world. For example, within the MEGAPOLI project, the global forecasts are provided by the MATCH-MPIC (Max Planck Institute for Chemistry version) model. These provide boundary conditions for several operational European regional CWFSs. The ECMWF global CWF model provided chemical boundary conditions for the regional-scale European CWF models in the GEMS project. The global CWF ensemble to be constructed in the MACC project will update the regional model ensemble provided within the GEMS project.

Although using boundary conditions from global models should in principle improve predictive skill in regional models by providing more realistic temporal and spatial variability, they also can transfer biases and errors. Further improvements are therefore needed in the observing systems that provide information on the three-dimensional pollutant concentrations. For example, such improvements are needed to better quantify the long-range pollutant transport of Saharan dust to Southern Europe and Asian brown dust cloud to the US West Coast (Huang et al., 2010).

7.4 Data assimilation of chemical species

As near-real-time measurements of chemical concentrations are limited, one of the challenges in CWF is how to insert that data into the models to obtain the best initial conditions (e.g. the initial spatial concentrations of the relevant chemical

species) and to improve the quality of CWF. The way this data is inserted is called data assimilation. Powerful assimilation techniques may actually be more critical for achieving accurate forecasts than improvements in the model formulations, at least regarding the short-range forecasts (1–2 days) (e.g. Carmichael et al., 2008a). The implementation of the various chemical data assimilation methods in CWF models is therefore one of the crucial tasks in the improvement of regional CWF models.

The assimilation of meteorological data has traditionally been an essential part of weather forecasting. Different methods of data assimilation are used in NWP models: Newtonian nudging, optimum interpolation, regional four-dimensional data assimilation (FDDA), ensemble Kalman filter (EnKF), the three-dimensional variational (3DVAR) and four-dimensional variational (4DVAR) data assimilation. In CWF, EnKFs and 4DVAR are most commonly used. Although beyond the scope of this paper to provide a description of these methods, we provide a few remarks relevant for data assimilation into CW models.

In both the 4DVAR and EnKF approaches, the difference between observed and model values is measured by a weighted sum of squares, where the weights are constructed from several covariance matrices. These matrices reflect uncertainties in both data and model, and at least some of them have very large dimensions. Ensemble methods circumvent the intractability of large covariance matrices by approximating them by an ensemble of model states (in the CWF case, these are usually states of the CWF model).

The EnKF advances each member of an ensemble one time step ahead. Then, an EnKF updating formula is applied, using observed data and covariance matrices approximated by low-rank sample covariance matrices defined by the ensemble. The classical EnKF update formula is based on assumptions of an unbiased model and the error distribution being Gaussian. In real-life applications, nonlinearity of chemical reactions causes departures from the Gaussian, whereas deficiencies in the model and errors in the inputs contribute to the bias (one typical example is nightly values of ozone in some models). The departures from the EnKF assumptions are much larger in CWF models than in NWP models. These issues and other types of filters are studied in Hanea et al. (2007).

A substantial difference between data assimilation in NWP models and CWF models is due to different types of model equations. In CWF models, stiff differential equations with forcing terms from meteorological and emission inputs make the model quickly converge from any reasonable initial conditions to a stable solution. Thus, in off-line CWF models, improvement of initial conditions by means of data assimilation brings only a limited improvement in the forecast. The same issue causes loss of spread in ensembles generated by perturbations of initial conditions. The sample covariance matrices generated by the ensemble become ill-conditioned, and covariance inflation or similar methods have to be used to avoid divergence of ensemble filters (Eben et al., 2005; Constantinescu et al., 2007).

Another challenge for the use of data assimilation in CWF models is that the number of chemical species in the models varies from tens to hundreds). A key issue is choosing which chemical species to optimize in order to provide the best results of the target forecast species. In variational methods, one may select a receptor location and investigate which variables or parameters are responsible for changes and errors of the model at the receptor. These methods (called adjoint sensitivity analysis or receptor-oriented approach; Carmichael et al., 2008a) may be used to select state variables for data assimilation. Also, short-lived species and radicals are usually not subject to optimization.

The difficulties mentioned above are the main reasons why data assimilation in CWF models has grown slowly relative to NWP modelling. Operational forecasting with incorporated data assimilation is occurring for the EURAD model and for RCG (only ozone maxima), and research-based studies have also been performed for LOTOS-EUROS, MATCH, RCG and SILAM. Most of the operational CWF models are routinely initialized using concentrations of species obtained from the forecasts of the previous day, with no regard to the observations. In order to make data assimilation more beneficial for the forecast, other parameters than initial conditions should be optimized, too, and the assimilation could be continued after the initialisation phase. Emission rates are the first candidate for optimization, but photolysis rates and deposition rates also may come into consideration (Hanea et al., 2004). However, stability and validity (from the point of view of chemistry or emission modelling) of such corrected parameters has to be checked in order to avoid artifacts.

Instead of optimizing initial conditions and other parameters for the operational forecasts, more sophisticated data assimilation methods can be applied. A fast-growing research area is inverse modelling of emissions using adjoint methods and 4DVAR. Although it is being used mainly in global modelling for monitoring atmospheric constituents (e.g. Kopacz et al., 2010), its benefit to forecasting has also been demonstrated (e.g. Elbern et al., 2000, 2007 for the EURAD model). In the US, the adjoints of the global model GEOS-Chem (Henze et al., 2007) and mesoscale models STEM (Sandu et al., 2005) and CMAQ (Hakami et al., 2007) have been developed. Adjoint modelling methods have been briefly reviewed in Table 13.

Research on both inverse modelling and data assimilation has been boosted by the availability of satellite-retrieved measurements (e.g. Chai et al., 2009). Global spatial coverage, better representativeness of the measured area and gradually improving resolution are the main virtues of these data, whereas censoring by clouds, relatively poor time resolution (e.g. two times daily over one spot) and inaccuracies of the retrieval process are the main drawbacks. Satellite instruments can also provide information that is largely complementary to that obtained from in-situ measurements. An

overview of European research on remote-sensing of tropospheric constituents is given by the ACCENT-TROPOSAT-2 (Atmospheric Composition Change the European Network – Use and Usability of Satellite Data for Tropospheric Research) report (Burrows and Borrel, 2009). We confine ourselves here to some general remarks related to CWF.

The satellite-based abundance data, which enter a data assimilation system, are most commonly integrated over the whole atmospheric column, although vertical profiles are also provided in some cases (e.g. the IASI instrument – Infrared Atmospheric Sounding Interferometer). Tropospheric columns are derived from total columns; one then has to address the generally poorer sensitivity of satellite observations to concentrations in the lower troposphere. Cloud cover has to be estimated, as well as other meteorological variables. For example, air-mass factor (the ratio between the retrieved slant column and the atmospheric vertical column) is needed for knowing the absorption of the light path through the atmosphere. As a result, satellite columns are a result of a complicated retrieval process leading from the observed spectra to a vertical column density. The uncertainty of the retrieval process therefore needs to be quantified for successful data assimilation.

In the future, real-time regional-scale CWF models may use data assimilation of the vertical distribution of chemical species using the vertical profile from a global CW model as a first guess. For example, a global CWF model TM (http://www.knmi.nl/~velthove/tm.html) is used in retrieving NO_2 column from the OMI instrument in the near-real-time service TEMIS (Tropospheric Emission Monitoring Internet Service, www.temis.nl) of the European Space Agency.

7.5 Improved understanding and parameterization of physical processes

The improvements required for the understanding and parameterization of subgrid-scale physical processes for CWF include at least two emerging areas. The first area is the accuracy of meteorological parameters (e.g. atmospheric boundary-layer structure, velocity, temperature, turbulence, humidity, cloud water, precipitation) within NWP models or in meteorological pre-processors (e.g. Fisher et al., 2006). The second area is the description of the interactions of chemical species in the atmosphere (e.g. clouds, radiation, removal processes, chemical reactions, aerosol formation and dynamics) within CWF models themselves.

Areas of necessary NWP model improvement include the overall treatment of complex terrain and rough surfaces (e.g. for urban areas), turbulence closure and mesoscale convection. The description of complex terrain and mesoscale circulations can be of crucial importance in CWF models (e.g. Millan et al., 1996; Gangoiti et al., 2001; Dayan and Levy, 2002; Dayan and Lamb, 2005). Among the most challenging cases for CWFSs to predict are episodes of high pollutant concentrations, which commonly occur with low winds and stable stratification, sometimes in complex terrain (e.g. Kukkonen et al., 2005a, b). These situations create problems for current methods and models to realistically reproduce meteorological input fields. In particular, the currently used NWP models may have severe problems in forecasting the occurrence and strength of strong ground-based temperature inversions (Pohjola et al., 2004; Rantamäki et al., 2005; Kukkonen et al., 2005a, b). A gap has thus emerged between modern understanding of boundary-layer physics and the limited applicability of boundary-layer schemes in operational CWF models.

As most of the pollutants are dispersed within the boundary layer, the mechanisms controlling concentrations substantially depend on the turbulence and the boundary-layer height. The temporal and spatial variations of the boundary-layer height and the entrainment processes at the top of the boundary layer lead to the infiltration of pollutants from the boundary layer to the free troposphere and, vice versa, to the intrusion of some chemical compounds (e.g. ozone) from the upper-atmospheric layers down to the surface. Physical processes controlling the boundary-layer height and the turbulent entrainment are therefore of crucial importance for CWFSs. Some of the important physical processes at the top of the boundary layer (e.g. Zilitinkevich et al., 2007) are still insufficiently understood, such as turbulent entrainment in rapidly deepening convective boundary layers and non-steady interactions between the stable boundary layers and the free flow. In the presence of cloud-topped boundary layers, the mixing and dispersion of gases is not always reflected well in traditional parameterization models, in which clouds usually suppress dispersion by diminishing solar irradiance. Due to the reduced vertical diffusion caused by clouds, precursors tend to remain near the ground level. As a consequence of the non-linearity of many chemical reactions, these concentration gradients can lead to different chemical reactions.

Most of the operational CWF models use simplified wet deposition schemes based on two-dimensional surface precipitation intensity data; however, on-line integrated models (e.g. Enviro-HIRLAM) are allowed to realise more comprehensive schemes using fully three-dimensional real-time cloud characteristics. One of the challenges in this emerging area is to improve the quality of the simulation of cloud processes and precipitation forecasts within NWP models. Increasing computational power makes it possible to decrease the horizontal and vertical grid spacings of the models. As the physical parameterization is dependent on the resolution of a prediction model, some adjustments of parameterizations have to be made when the resolution is increased.

Piriou et al. (2007) presented an approach in which the grid-scale budget equations of parameterization used separate microphysics and transport terms. This separation is used both as a way to introduce into the parameterization a more explicit causal link between all involved processes and as a vehicle for an easier representation of the memory of

convective cells. Piriou et al. (2007) argued that future results could be improved by using more complex microphysics (e.g. prognostic liquid, ice, rain, snow), getting closer to that of a cloud-permitting model, and relaxing the small-area assumption. As discussed by Roebber et al. (2004), however, the interpretation of forecasts at cloud-permitting resolutions becomes different than at larger grid spacings, complicating – if not offsetting – advantages of going to smaller scales.

In the future, a possible perspective will be to unify the convection parameterization, using a single equation set at grid-scale and a single microphysical package. As an example, Gerard (2007) has introduced microphysics and transport advective scheme equations into a scheme using more complex prognostic microphysics, area fraction, and vertical velocity with encouraging results. Gerard (2007) developed a package that aims at efficiently combining resolved and subgrid condensation at all resolutions, in particular in the range between 10 and 2 km, where deep convection is partly resolved and partly subgrid.

Knowledge of the emissions of relevant organic species and their atmospheric chemistry limits the understanding of secondary organic aerosols, which are of importance for both air quality and climate change (e.g. Monks et al., 2009). Correspondingly, the models for aerosol formation and dynamics need to be implemented into CWF models, and the chemical mechanisms used in CWF models should be substantially improved to be able to simulate sufficiently accurately such processes.

Combined models have already been developed for chemistry transport modelling and aerosol processes, including the size distributions and chemical speciation (e.g. Vignati et al., 2004; Gross and Baklanov, 2004; Dusek et al., 2006; Pohjola et al., 2007; Hussein et al., 2007; Medina et al., 2007; Langmann et al., 2008). The aerosol processes include the growth and nucleation processes, and the transport and deposition pathways of the aerosols. Furthermore, as aerosol dynamics models (this term is used here as a synonym to aerosol process models) are important tools to investigate both the direct and the indirect effects on climate, aerosol-radiation-cloud interactions are important processes that need to be treated in the models (Ramanathan et al., 2001; Rosenfeld et al., 2008; Levin and Cotton, 2009). Several of these processes require direct coupling of the meteorological and chemical weather models.

These processes need to be included to achieve a comprehensive representation of the atmosphere. State-of-the-art aerosol modules include a sectional representation of the size and chemical composition distribution functions, as well as aerosol microphysical processes. However, the available emission databases do not currently provide sufficient detail for executing such combined dispersion and aerosol process models over extensive regions. This means that estimates of the sectional emissions that are needed as input for the detailed models have to be mostly based on indirect information.

Improvements in three-dimensional wet-deposition mechanisms within and below clouds for interacting different chemical gas and aerosol species are needed. Improvements in the parameterization of pollutant emissions removal processes in the subgrid-scale are also required. Any removal that may occur near the emission source (on a scale of tens to hundreds of meters) is usually beyond our present capability. Pace (2005) discuss the need to treat the subgrid-scale, near-source fugitive dust emissions removal by settling and impaction on surface cover.

7.6 Better evaluation of CWF models with data

Model evaluation is fundamental to build confidence in the models and their specific applications. Nevertheless, the evaluation of models by comparison with measured data also has to advance the model performance, rather than solely characterize whether a simulation is successful or not (e.g. Gilliland et al., 2008). The comparisons should use as broad and diverse set of measured data as possible. Currently the evaluation of chemical weather models is mainly based on the comparison of measured and simulated concentration levels at ground level, although satellite data is also commonly used. Clearly, a data comparison only based on one vertical level does not assure a proper simulation of the state of the atmosphere. Whenever possible, vertical profiles of air pollutants, such as MOZAIC data (Kalabokas et al., 2007), should be included in the evaluation procedure.

Moreover, the performance of models is usually evaluated only for a limited number of pollutants, such as NO_x, O_3 and PM_{10}, which are the ones measured routinely at most monitoring networks. An ideal comparison would be based on the analysis of a sufficiently large number of pollutants, for the selected period of time (or periods) and location (or locations). Clearly, monitoring supersites (or their networks), such as the EPA's particulate matter supersites (e.g. Sioutas et al., 2004; Stanier and Solomon, 2006) or the Mace Head Research Station supersite in Ireland (e.g. Cape et al., 2000; Heard et al., 2006), are useful for this kind of evaluation and could potentially allow for an evaluation of the model capabilities to simulate various physical-chemical processes.

A special concern for the evaluation of operational CWF models is the availability of near-real-time meteorological and air quality data. Efforts to deliver near-real-time data (centralised in a common and accessible database) have been made within the GEMS project, which fits into the current WMO (World Meteorological Organization) activities. However, there is still a substantial amount of work to be done in this area; fast mechanisms need to be developed, implemented and tested to access the data and to evaluate the CWF models.

Currently, the evaluation of particulate matter in models commonly uses mostly the measurements of PM_{10} and, only to a smaller extent, $PM_{2.5}$, and size- and chemically-resolved PM data. However, the evaluation of models in

Europe should in the future focus on $PM_{2.5}$ (or PM_1) instead of PM_{10}, as it is more relevant from a health perspective. Clearly, more focus should also be on transferred to other PM measures instead of particulate mass, such as particle number concentrations, ultrafine particles, and chemically resolved size distributions. Due to the new European legislation, $PM_{2.5}$ monitoring data will be extensively measured in the European Union, and the new monitoring network needs to be evaluated.

Furthermore, the understanding and evaluation of the chemical components of particulate matter is needed to ensure that the model predictions are right for the right reasons and to close the gap between modeled and measured concentrations of PM. For instance, the evaluation of only the total PM_{10} concentration may not reveal serious shortcomings in the treatments of the PM components in the model. Moreover, size-resolved PM data are crucial to reduce uncertainties in our understanding of the modelling of the emissions, dispersion and transformation of PM. Aerosol chemistry and process modules are needed to evaluate the aerosol components. In that case, the model evaluation does not need to be restricted to only PM_{10} and $PM_{2.5}$.

Clearly, measurements routinely carried out at air pollution networks can only be used for model evaluation in case of a limited set of chemical components and measures. Species of interest for model comparison are not necessarily measured (e.g. peroxyacetyl nitrate (PAN), aldehydes, actinic flux, turbulent flux, other particulate matter measures except for PM masses), and the measurement locations are not always representative of the regional background air (e.g. the station could be located downwind of an urban area). In addition, the vertical profiles of concentrations are typically not measured.

More emphasis should also be given to the systematic evaluation of the spatial representativeness of the monitored and simulated data. Clearly, when comparing model predictions to measured data we compare one spatially and temporally averaged value (a predicted one) to another one (the measured one). However, in general, the averaging or representative space and time scales are not the same (Kang et. al., 2007).

The model evaluation against observed data requires statistical analysis that should provide information about the ability of the model to predict the observed values and type of errors (systematic or unsystematic). Generally, the statistical analysis contains a computation of a set of parameters and measures. It is possible to define various subsets of such statistical parameters that can fairly well represent the various aspects of the performance of the model (e.g. correlation coefficient, fractional bias, root and normalized mean square errors). However, these statistical quality indicators should be accompanied by other methods – time series and scatterplots could be an important complement to the above statistical indicators. Clearly, parameters that reflect the capability to simulate concentration peaks should also be taken into consideration in CWF.

Besides the comparison of model results to data, several other steps should be considered to ensure model quality. These include model sensitivity tests, model intercomparisons and uncertainty analysis (e.g. Borrego et al., 2008). Several international model intercomparison exercises have been and are currently ongoing within the MEGAPOLI and MACC projects, CityDelta and EURODELTA (Sect. 5.3), various COST Actions, AQMEII and FAIRMODE (Forum for AIR quality MODElling). Such intercomparison exercises are useful to identify the strengths and weaknesses of models, and to show the strategies to improve their performance. Their important role in collecting intensive data during special field campaigns to assist diagnostic evaluation of model processes should also be acknowledged.

7.7 Model ensembles

Ensemble forecasting has been a key area of traditional meteorology during the last few decades (Lewis, 2005). From the experience of operational meteorology, two major sources of forecast errors can be distinguished. In operational meteorology, the first source resides in the uncertainties of the initial meteorological conditions, as a result of the limited number and inaccuracies of available observations. The second source is the imperfection of the NWP models, resulting from limitations in the descriptions of physical processes, the finite spatio-temporal resolution of numerical models and the inability to explicitly resolve and simulate processes beyond the selected grid scale. As a consequence of these two sources of forecast errors, and due to instabilities of the flow itself, weather forecasts deteriorate as the forecasting period increases.

In addition to the accuracy of the initial conditions and the limitations of the numerical model, the forecast skill also depends on instabilities of the flow itself, as was already identified in the early works of Lorenz (1963, 1965). Simmons et al. (1995) note the difficulty in assessing a priori whether a forecast would be skillfull or unskillfull, using only a deterministic approach to weather prediction.

Two important benchmarks occurred in the implementation of operational ensemble prediction systems, at both ECMWF and NCEP (e.g. Palmer et al., 1993; Molteni et al., 1996; Tracton and Kalnay, 1993). These systems were focused on the perturbation of the initial conditions, following indications on the relative importance of the uncertainties in initial conditions compared to deficiencies in the model (e.g. Downton and Bell, 1988; Richardson, 1998). Ensemble forecasting continues to be an area of active research in NWP, specifically the design and composition of the ensembles (e.g. Kalnay, 2002).

Ensemble CWF is still an emerging area (e.g. Potempski et al., 2008). There are currently numerous well-evaluated CWFISs in Europe that are used both for research and

operational applications in direct support of decision making. However, any of these single modelling approaches bears inherent uncertainties, both originated from its formulation, the parameterization used, and from the input data used (e.g. meteorology, emissions, chemical rate constants). It is therefore desirable to enrich the information provided by the individual deterministic models with probabilistic information (e.g. Kukkonen et al., 2009c). The three key objectives of ensemble forecasting (e.g. Kalnay, 2002) are to (i) improve the forecast by ensemble averaging, (ii) to provide an indication of the reliability of the forecast, and (iii) to provide a quantitative basis for probabilistic forecasting. The spread of predictions in a collection of models can also be used as a measure of the model uncertainty (Vautard et al., 2006). The comparison of the predictions of model ensemble and those of the individual models can also give valuable insight on model performance, e.g. regarding model outliers for specific pollutants or conditions.

Compared to traditional weather forecasting using model ensembles, chemical weather ensemble prediction has a much shorter historical record. Early studies comprise works in the field of chemical weather forecasting (Delle Monache and Stull, 2003) and chemical transport modelling (Galmarini et al., 2004b, c). As in NWP, these studies have investigated both techniques based on the perturbation of single modelling systems (Mallet and Sportisse, 2006) and on a collection of results from different modelling systems (van Loon et al., 2007; Vautard et al., 2008). Unlike in meteorology, however, air quality is not primarily determined by initial conditions but rather is the result of a range of processes such as emissions, transport, deposition and chemistry that all provide tendencies with similar orders of magnitude. This state of affairs requires one to develop approaches that are more complex than the well-established techniques used in numerical weather prediction (e.g. Pinder et al., 2009).

The forecasts obtained by processing the ensemble of models (for instance, taking the median of all values in each grid-point) can, in many cases, perform better than any single model. Riccio et al. (2007) have proposed a theoretical basis in the case of dispersion, providing some justification to the relatively better performance of the median of models. Recently, Galmarini et al. (2010) evaluated various ensemble atmospheric dispersion simulations for the ETEX-1 tracer experiment case. They analyzed on one hand the so-called multi-model prediction systems that rely on model simulations produced by different CTM's using meteorological data from potentially different weather prediction systems, on the other hand prediction systems running a single atmospheric dispersion model with the ensemble weather prediction members.

The current operations in the GEMS and MACC projects have used a more elaborate ensemble technique, based upon the differential weighting of the individual models according to their skill over the last few days. However, a long-term improvement in CWF performances will be based on the improvement of individual models and their representation of dynamical, physical and chemical processes.

8 Conclusions

What do we see in the future for CWF models? To summarize this paper, we focus on two challenge areas: the large number of chemical species and processes, and communicating uncertainty.

First, although a relatively new field, CWF is developing quickly, touching upon research, development, and operational forecasting. An analogy with weather forecasting can be useful to demonstrate the challenges ahead. Although CTMs can be coupled to NWP models either off-line or on-line at present, a scientific perspective of CWF would argue for an eventual migration from off-line modelling (where the CTM is run after the NWP model run is completed) to on-line modelling, allowing coupling and integration of the physical and the chemical components of CWFISs. Such a future is not hard to imagine, given similar trends of Earth system modelling, for example. Specifically, better and more complete representations of physical and chemical processes and interactions in the models are needed. When compared to weather forecasting, CWF still has a long way to go. Despite the nearly 50-yr lead that NWP has over CWF (e.g. Harper et al., 2007), CWF models have other challenges that inhibit as rapid a progress.

A key challenge appears to rather be the dimensionality and complexity of the problem itself. For example, the traditional set of prognostic state variables in weather forecasting (e.g. temperature, wind, precipitation) expands to hundreds of prognostic variables because of the extensive number of chemical species involved. In particular, resolving, simulating, and parameterizing processes is no longer limited to relatively well-known physical processes, but is compounded by a huge amount of both chemical and physical processes (e.g. interactions between species, emission, deposition, radiation). This simple fact has important ramifications for predictability, data assimilation, and ensemble prediction, where scientific and technological progress in CWF is slower than in traditional meteorology. Importantly, progress is also inhibited by the lack of or insufficient monitoring of many relevant species, the lack of sufficient chemical and aerosol measurements, and the lack of well-established monitoring data-exchange mechanisms, although several projects and initiatives are working to address these issues.

Second, as is evident from this review article, numerous well-evaluated operational CWFISs operate in Europe, addressing the needs of a large spectrum of users from governmental organizations to the individual citizen (e.g. Schluenzen and Sokhi, 2008; Karatzas and Kukkonen, 2009; Baklanov et al., 2010b; Balk et al., 2011). Despite these numerous activities, it is challenging to transfer the output from

CWF models for the end users in a form that is properly assessed and interpreted. Moreover, how do we interact with those users to provide the needed services? Through initiatives such as the GMES Atmospheric Service and its implementation projects GEMS, PROMOTE, MACC and PASODOBLE and the various relevant COST actions, such as COST 728, ES0602 and ES1004 scattered modelling initiatives and efforts – which are often national or regional in scale – can be integrated. Such integration also offers the possibility to move from deterministic forecasts of chemical weather to ensemble chemical weather prediction systems.

With the ability to assess and explore ensemble prediction systems comes the challenge in communicating probabilistic chemical weather forecasts. Again, many lessons can be learned from the weather forecasting community, who are actively facing such concerns with weather forecasts, in general (e.g. Board on Atmospheric Sciences and Climate, 2006; Novak et al., 2008; Morss et al., 2008; Rauhala and Schultz, 2009), and the communication of hurricane tracks, in particular (e.g. Broad et al., 2007). Although some user communities of weather information (e.g. industrial, agricultural, hydrological) are comfortable dealing with probabilistic forecasts, many chemical weather users are relatively new to this concept. Thus, much can be gained through a closer dialogue with relevant user communities (e.g. the so-called "end-to-end-to-end" approach described by Morss et al., 2005), and this communication can spawn future research opportunities.

Successful CWFIS services will also need to aggregate and integrate information and deliver it in a way that is comprehensible, user-friendly, timely, and reliable. International activities such as the WMO Global Atmospheric Watch Urban Research Meteorology and Environment (GURME) project can assist in these efforts. As a first step, the European CWF portal created by the COST ES0602 action (Balk et al., 2011) attempts to integrate existing chemical weather forecast and information solutions offered by numerous institutions within Europe. This portal provides a direct gateway to the individual resources and is intended to complement and support other European initiatives such as the GMES Atmospheric Services.

Appendix A

Abbreviations and acronyms used in this article

ACCENT-TROPOSAT-2	Atmospheric Composition Change the European Network – Use and Usability of Satellite Data for Tropospheric Research
ACM	Asymmetric Convective Model
aero3	3rd generation CMAQ aerosol module
ALADIN	Aire Limite Adaptation Dynamique INitialisation
ALADIN-CAMx	Comprehensive Air-quality Model with extensions based on ALADIN-Austria forecast data
AM&WFG	Atmospheric Modeling and Weather Forecasting Group
ANA	Atmospheric mesoscale Numerical pollution model for urban and regional Areas
AOD	Aerosol Optical Depth
AROME	Applications of Research to Operations at Mesoscale
ARPA	Italian Regional Environmental Protection Agency
ARW	Advanced Research WRF
AURAMS	A Unified (multiple-pollutant) size- and chemical composition-resolved, episodic, Regional Air-quality Modelling System
AUTH-NKUA	Aristotle University of Thessaloniki – National and Kapodistrian University of Athens
AQA	Air-Quality model for Austria
AQMEII	Air-Quality Modeling Evaluation International Initiatives
BEIS3	Biogenic Emission Inventory System
BIOEMI	BIOgenic EMIssion model
BNMVOC	Biogenic Non-Methane Volatile Organic Compound
BOKU	the University of Natural Resources and Applied Life Sciences in Vienna
CAC	tropospheric Chemistry Aerosol Cloud transport model
CALGRID	California Grid Model
CAM3	Community Atmosphere Model v.3
CAMx	Comprehensive Air-quality Model with extensions
CAMx-AMWFG	Comprehensive Air-Quality Model with Extensions – The Atmospheric Modeling and Weather Forecasting Group
CBM-IV	Carbon Bond Mechanism IV
CCN	Cloud Condensation Nuclei
CECILIA	Central and Eastern Europe Climate Change Impact and Vulnerability Assessment
CITYZEN	megaCITY – Zoom for the Environment
CMAQ	United States Environmental Prediction Agency, Community Multiscale Air-Quality Model
COSMO	Consortium for Small Scale Modeling
COST	European Cooperation in Science and Technology
CTM	Chemistry-Transport Models
CWF	Chemical Weather Forecasting
CWFIS	Chemical Weather Forecasting and Information System
CWFS	Chemical Weather Forecasting System
DDM	direct decoupled method
DEHM	Danish Eulerian Hemispheric Model
DERMA	Danish Emergency Response Model of the Atmosphere
DMAT	Dispersion Model for Atmospheric Transport
DMI	Danish Meteorological Institute
DMS	DiMethyl Sulfide
DREAM	Danish Rimpuff and Eulerian Accidental release Model
DWD	German Weather Service

ECMWF	European Centre for Medium-Range Weather Forecasts
ECWFP	European open-access Chemical Weather Forecasting Portal
EDGAR	Emission Database for Global Atmospheric Research
EEA/MDS	European Environment Agency/Model Documentation System
EEM	EURAD Emission Module
EMEP	European Monitoring and Evaluation Programme
EMIMO	EMIssion MOdel
ENEA	Ente per le Nuove tecnologie, l'Energia e l'Ambiente
EnKF	Ensemble Kalman Filtering
Enviro-HIRLAM	Environment-HIgh Resolution Limited Area Model
EPA	US Environmental Protection Agency
ESCOMPTE	Expérience sur Site pour COntraindre les Modéles de Pollution atmosphéique et de Transport d'Emissions
Esquif	Étude et Simulation de la QUalité de l'air en lle de France, a synthesis of the Air Pollution Over the Paris Region
ETEX	European Tracer Experiment
EUCAARI	European Integrated project on Aerosol Cloud Climate and Air-Quality Interactions
EU-ESA	European Space Agency
EuMetChem	European framework for on-line integrated air-quality and meteorology modelling
EUMETNET	Network of European Meteorological services
EURAD	European Air Pollution and Dispersion Model
EUSAAR	European Supersites for Atmospheric Aerosol Research
EQSAM	The Equilibrium Simplified Aerosol Module
EZM	European Zooming Model
FAIRMODE	Forum for AIR-quality MODelling
FARM	Flexible Air-quality Regional Model
FDDA	Four-Dimensional Data Assimilation
FMI	Finnish Meteorological Institute
FRP	Fire Radiative Power
GATOR-GCMOM	Gas, Aerosol, Transport, Radiation, General Circulation, Mesoscale, and Ocean Model
GCM	General Circulation Model
GEIA	Global Emissions Inventory Activity
GEMS	Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data
GEOmon	Global Earth Observation and Monitoring program
GEOSS	Blobal Earth Observation System of Systems
GFS	Global Forecast System
GME	Global Model of DWD (DWD – German Weather Service)
GMES	Global Monitoring for Environment and Security
GOME2	The Global Ozone Monitoring Experiment-2
GRADS	GRid Analysis and Display System
GURME	World Meteorological Organization Global AtmospherIc Watch, Urban Research Meteorology and Environment
HARMONIE	Hirlam Aladin Research on Meso-scale Operational NWP in Euromed
HIRLAM	HIgh Resolution Limited Area Model
IASI	Infrared Atmospheric Sounding Interferometer
IDV	Integrated Data Viewer
IFS	Integrated Forecast System (ECMWF)
IIASA	International Institute of Applied System Analysis
ICLAMS	Integrated Community Limited Area Modeling System
IPCC	Intergovernmental Panel on Climate Change
IPSL	Laboratoire de Météorologie Dynamique
ISORROPIA	Greek aerosol module
KPP	Kinetic Pre-Processor
LAEI	London Atmospheric Emission Inventory database
LOTOS-EUROS	LOng Term Ozone Simulation-EURopean Operational Smog model
LRTAP	Long-range Transboundary Air Pollution
LSODE	Livermore Solver for Ordinary Differential Equations
MACC	Monitoring Atmospheric Composition and Climate
MADE	Model Aerosol-Dynamics model for EURAD
MARS	Model for the Atmospheric Dispersion of Reactive Species
MATCH	Multi-scale Atmospheric Transport and Chemistry Model
MDS	Model Documentation System
MEGAN	Model of Emissions of Gases and Aerosols from Nature
MEGAPOLI	Megacities: emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation
MELCHIOR	Modele Lagrangien de Chimie de l'Ozone a l'échelle Régionale
MEMO	MEsoscale Model
MINNI	Modello Integrato Nazionale a supporto della Negoziazione Internazionale sui temi dell-inquinamento atmosferico
MIRAGE	Model for Integrated Research on Atmospheric Global Exchanges
MM5	Fifth Generation PSU/NCAR Mesoscale Model
MM5-CAMx	Fifth Generation PSU/NCAR Mesoscale Model – Comprehensive Air-quality Model with extensions
MM5-CHIMERE	Fifth Generation PSU/NCAR Mesoscale Model – CHIMERE

MM5-CMAQ	Fifth Generation PSU/NCAR Mesoscale Model – Community Multiscale Air-Quality Model
MOCAGE	Modéle de Chimie Atmosphérique à Grande Echelle, Model of Atmospheric Composition at Large Scales
MODIS	Moderate Resolution Imaging Spectroradiometer
MOPITT	Measurements Of Pollution In The Troposphere
MPIC	Max Planck Institute for Chemistry
MRF	Medium Range Forecast model
M3Dry	dry deposition velocity scheme
NAME	Numerical Atmospheric-dispersion Modelling Environment
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
NETCDF	NETwork Common Data Form
NMM	Nonhydrostatic Mesoscale Model
NMVOC	Non-Methane Volatile Organic Compounds
NOAA	National Oceanic and Atmospheric Administration
NWP	Numerical Weather Prediction
NWP-Chem-liquid	thermodynamic equilibrium model
OFIS	Ozone Fine Structure model
OMI	Ozone Monitoring Instrument
OML	point source model
OPANA	Operational version of Atmospheric mesoscale Numerical pollution model for urban and regional Areas
OSPM	Operational Street Pollution Model
PALM	computational fluid dynamics software
PASODOBLE	Promote Air-Quality Services integrating Observations – Development Of Basic Localised Information for Europe
PAVE	Package for Analysis and Visualization of Environmental data
PBL	Planetary Boundary Layer
PM	Particulate Matter
PNNL	Pacific Northwest National Laboratory
POP	Persistent Organic Pollutant
PPM	Piecewise Parabolic Method
PREVAIR	Air-Quality forecasts and observations in France and Europe
PROMOTE	PROtocol MOniToring for the GMES Service Element
PSU/NCAR	Pennsylvania State University/National Center for Atmospheric Research
RACM	Regional Atmospheric Chemistry Mechanism
RADM	Regional Acid Deposition Model
RAMS	Regional Atmospheric Model System
RCG (REM3-CALGRID)	Regional Eulerian Model – California Grid Model
REMEST	a nonhydrostatic mesoscale meteorological model
RETRO	Reanalysis of the TROpospheric chemical composition
RIU	Rhenish Institute for Environmental Research
RIVM	National Institute for Public Health and the Environment
SAPRC99	chemical mechanism developed at the Statewide Air Pollution Research Center in Riverside, California
SILAM	Air-Quality and Emergency Modelling System
SMHI	Swedish Meteorological and Hydrological Institute
SMOKE	Sparse Matrix Operator Kernel Emissions model
SNAP	Selected Nomenclature for Air Pollution
SOA	Secondary Organic Aerosols
STEM	Sulfur Transport and dEposition Model
STOCHEM	UK Meteorological Office Chemistry-transport Model
STRACO	Soft TRAnsition Convection
SURFPRO	SURrface atmosphere interFace PROcessor
TA	Temperature Anomaly
TEMIS	Tropospheric Emission Monitoring Internet Service
TESSEL	Tiled ECMWF Scheme for Surface Exchanges over Land
THOR	an integrated air pollution forecast and scenario management system
TM5	Tracer Model 5
TNO	the Netherlands Organisation for Applied Scientific Research, Utrecht, Netherlands
TRAMPER	Tropospheric Realtime Applied Meteorological Procedures for Environmental Research
TRANSPHORM	Transport related Air Pollution and Health Impacts – Integrated Methodologies for Assessing Particulate Matter
TSP	Total mass of Suspended Particles
TUV	Tropospheric Ultraviolet-Visible model
UBM	Urban Background Model
UM	Unified Model
USGS	United States Geological Survey
VOC	Volatile Organic Compounds
WMO	World Meteorological Organization
WRF-Chem	Weather Research and Forecast model coupled with Chemistry
YSU	Yonsei University
3DVAR	three-dimensional variational assimilation approach
4DVAR	3DVAR with assimilating data in time

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