

**MODELLING OF TRANSPORT AND TRANSFORMATION OF
AIR POLLUTANTS
– PRACTICAL INFORMATION FOR MODEL USERS**

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1. Introduction

Growing interest in air quality management and forecasting has resulted in the development of tools for modelling of air pollutants' transport and transformation. There are several state-of-the-art air quality models that can be applied for different air pollution problems.

Model users, apart from having basic knowledge on meteorological and chemical processes in the atmosphere, should be aware of some technical aspects of the air quality modelling and the air quality models' structure. This paper describes some of the practical issues concerning model configuration, required input and possible dependencies between different modules.



Figure 1.1. Air quality model calculates the evolution of concentration fields of air pollutants, in subsequent timesteps (C_i – concentration of “i” modelled species).

2. Applications - timescale and spatial scale of air quality problems

The type of the air quality model, suitable for specific purpose, depends on the spatial and temporal scale of the analysed problem. Air quality processes can be considered as short term (hours to days) or long term (season to years). As common examples of short term AQ modelling, following applications can be mentioned:

- operational air quality forecast;
- case studies of severe air pollution episodes;
- emergency response / risk assessment systems;

Long term simulations may support problems related to air quality policy e.g.:

- air quality assessment;
- scenarios of emission reduction and regions development;
- regulatory purposes;
- climate and climate change modelling.

Spatial scale of the simulated case is closely connected with the temporal scale of the reproduced processes. Global air quality models are usually used for long-term applications like climate and climate change modelling (e.g. IPCC), investigating the long-range and transcontinental transport of air pollutants (e.g. HTAP) or spatial distribution of the important trace species in the atmosphere analysis (e.g. O_3 , CO_2).

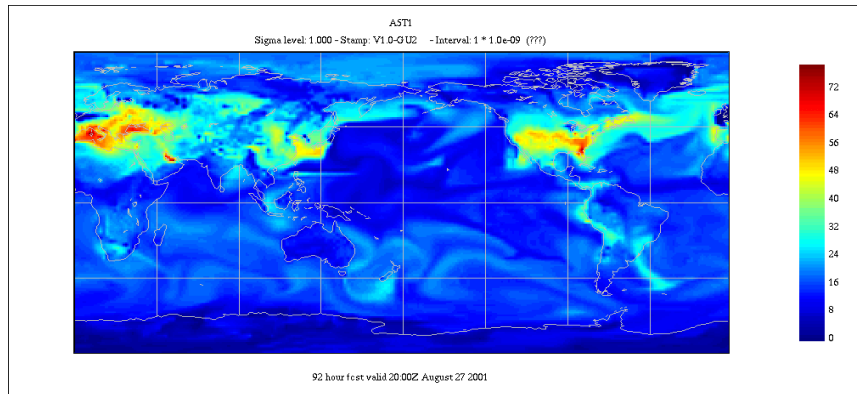


Figure 2.1. An example of global computational grid (uniform resolution). The picture shows surface ozone concentration calculated with GEM-AQ model (simulation performed by Warsaw University of Technology).

Regional and meso-scale air quality problems are connected with both: long- and short term applications. All examples given above for temporal scales (in spite of regulatory procedure) can be applied to regional scale.

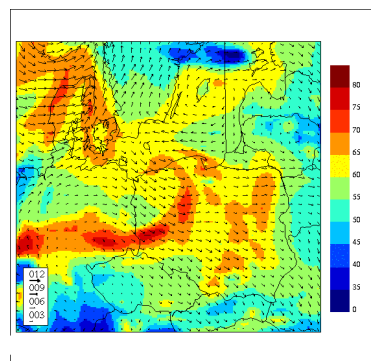


Figure 2.2. An example of computational grid for the regional scale. The picture shows surface ozone concentration (ppb) and wind field over Poland (simulation performed by Warsaw University of Technology).

Local scale problems (usually modelled at the resolution ranging from hundred meters to ~5 kilometres) are connected with the dispersion in street canyons, distribution of air pollutants over urban areas and with modelling of dispersion near large point sources (e.g. for regulatory purposes).

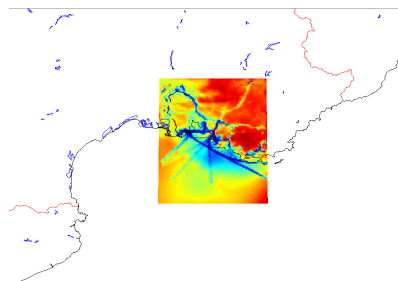


Figure 2.3. An example of computational grid for local scale. The picture shows surface ozone concentration near Marseille (simulation performed by Warsaw University of Technology in the frame of ESCOMPTE Modelling Exercise).

3. Air Quality Model (AQM) structure

Numerical air quality models are based on the mathematical description of transport and transformation of the trace species in the atmosphere. The transport equation calculates the changes in concentrations fields for each modelled pollutant. These changes are due to advective transport, vertical diffusion, and different sources and sinks, such as chemical transformation, anthropogenic and natural emissions, dry deposition and wet removal.

The complexity of the model depends on the number of pollutants and chemical reactions as well as number of physico-chemical processes treated. These processes are parameterised based on certain assumptions and simplifications. Choosing specific air quality model, the user should be aware of its limitations, especially while using simple models.

3.1. Sub-modules

The processes included in AQ model (described on Fig.3.1) are grouped into following modules:

- 1) Meteorological processor, provides 3D winds fields, temperature, humidity, and other meteorological parameters. Described processes may include interactions between surface and atmosphere, boundary layer dynamics and the exchange between the boundary layer and free troposphere. More information on the types of meteorological processors can be found in the paper by Łobocki included in this issue.
- 2) Emissions module
Emission inventory must be prepared with care, since reasonably accurate emission fluxes are necessary to obtain realistic concentration fields and deposition fluxes of modelled air pollutants. List of chemicals that should be included in the emissions inventory depends on the formulation of chemical module. More information on emission inventories and emission data processing will be given in the next section.
- 3) Air quality module, describes chemical reactions and removal processes for modelled species.

Chemical mechanism

All chemical mechanisms included in air quality models are approximations of the real atmospheric processes. Chemicals and chemical reactions are condensed into a smaller number of representative species and reactions. In most of the advanced models, several tens species combined with one to two hundreds of chemical reactions are included.

Advanced, state-of-the-art chemical mechanisms should contain gas phase chemical reactions module, aqueous phase chemical reactions module, aerosol module and heterogeneous chemistry module. Modelling of aerosol formation is very complex and should take into account size distribution and aerosols' chemical composition. The processes treated include nucleation, condensation, coagulation, dry deposition and wet removal. However, physics and chemistry of aerosols are yet not well understood.

Removal processes

Dry deposition is the transport of gaseous and particulate pollutants from the atmosphere to the surfaces in the absence of precipitation. The term "wet deposition" covers natural processes by which the air pollutants are scavenged by clouds, rain or snow and are deposited at the surface.

Dry deposition sink is commonly considered in models by calculating the dry deposition flux based on the surface layer concentration and the dry deposition velocity. Dry deposition velocity describes the efficiency of the turbulent transport, molecular diffusion (and sedimentation of aerosols) and uptake by the vegetation cover, soils, snow and ice sheets and water surface. Dry deposition process depends on the atmospheric stability, chemical properties of the modelled gases and particles and specific properties of the contacting surface.

Aerosol and gas species scavenged inside the cloud can be incorporated into raindrops and cloud drops via nucleation scavenging and dissolution (in-cloud scavenging). Main process of below-cloud scavenging of soluble species is dissolution into the falling raindrops.

Removal processes can be described on different levels of complexity. Simple parameterizations are based on constant values of parameters characterizing intensity of removal processes such as dry deposition velocity and scavenging coefficient for the modelled species. More complex parameterizations allows for calculation of this parameters in respect to meteorological situation and surface characteristics.

3.2. Coupling between meteorological and chemical module

As shown on Fig. 3.1, there are many dependencies between sub-modules of air quality model. The connection between meteorological parameters and chemical processes in the atmosphere is very close. The quality of meteorological data and accuracy of physical processes parameterisation are crucial for calculation of horizontal transport, vertical diffusion, chemical transformation and efficiency of dry and wet removal processes. Chemical reaction rates depend on temperature, pressure, and humidity. Solar radiation (solar zenith angle, cloudiness) impact on the intensity of photochemical transformation; reaction rates for aqua's phase chemistry depend on humidity and liquid water content. Also, meteorological conditions impact on emissions flux intensity, especially for species emitted from biogenic sources and also on efficiency of removal processes.

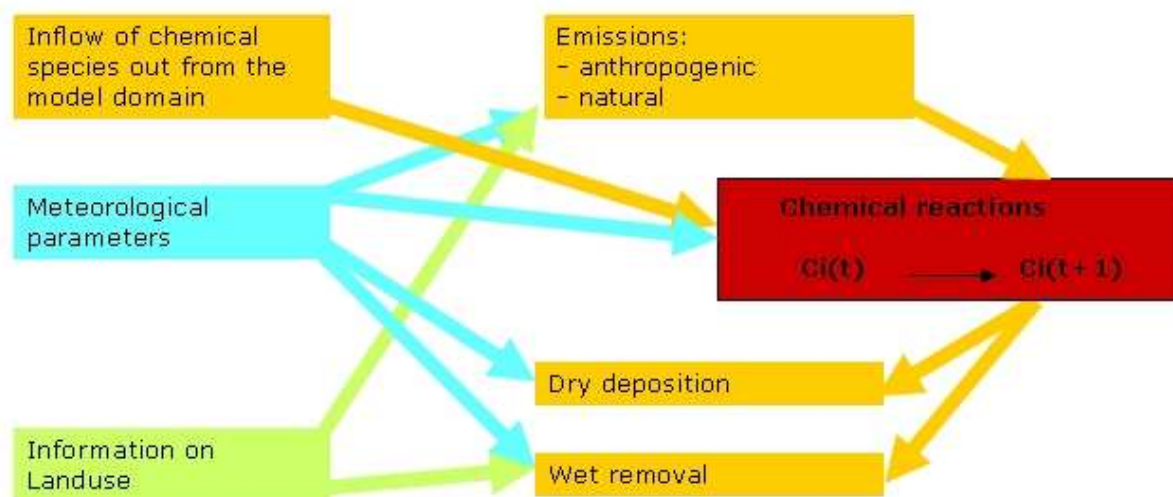


Figure 3.1. Simplified scheme of chemical module and possible dependencies between sub-modules:

- meteorological parameters influence intensity of emission, modify chemical reaction constants and impact on dry and removal processes;
- information on surface parameters and landuse modify natural and biogenic emissions and impact on dry deposition processes;
- emission and inflow through boundaries acts as sources;
- dry and wet removal processes acts as sink processes;
- chemical solver calculated changes in pollutants concentration caused by transformation in chemical reactions.

There are different methods of introducing meteorological information to air quality model. Simplified models use climatological approached (wind rose, classification of stability classes). At present time most of the models use sequential meteorological data. Several models read in and process measurement data with a built in diagnostic model. Other group of models can use 3D meteorological fields provided by mesoscale meteorological models.

Depending on the strategy of coupling between meteorological module and chemical module, models can be distinguished as off-line and on-line. In the off-line models, meteorological information is produced is separate simulation by meteorological model and than used in next simulation (in certain intervals – e.g. 1 hour) to drive chemical transport model. In the on-line model, the chemical module is incorporated into meteorological model, and calculations for meteorology and chemistry are performed simultaneously. Main advantage of the on-line models is

better quality of meteorological data and its availability of updated meteorological information in each computational timestep. Such approach allows to avoid errors due to spatial, vertical and time interpolation, and minimising inconsistencies connected with differences in geophysical and landuse data.

In modern climate model more advanced approach is used - the chemical information is transferred back to meteorological module, and changes in the chemical composition of the atmosphere impact radiation, and cloud formation processes (CCN). Such models are called interactive.

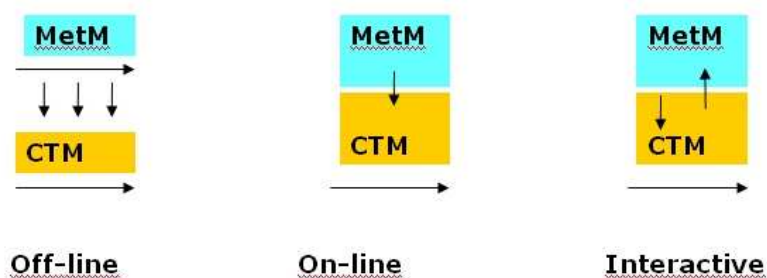


Figure 3.2. Coupling of meteorological and chemical transport models.

4. Data requirements and data sources

The typical structure of the air quality model, described above, determines input data requirement necessary to drive all incorporated modules.

Meteorological data

Depending on the complexity of the model several meteorological data might be required, e.g.:

- wind speed and wind direction;
- humidity;
- air temperature;
- surface pressure;
- solar radiation;
- cloudiness;
- precipitation.

These gridded fields may be generated using observations or provided by meteorological model. Most of AQ models include meteorological module that allows for processing measurements and computing required meteorological parameters. CALMET in CALPUFF¹ modelling system or AERMET in AERMOD² model are common examples of such pre-processors. Two types of measurements can be read into the model – surface measurements and vertical soundings. Surface data can be obtained from SYNOP reports (measurements from routine synoptic station); vertical soundings are coded in TEMP reports.

Many AQ models can be driven with gridded data produced by meteorological mesoscale models. At present, some models are available for scientific community as community models, e.g.: MM5³, often coupled off-line with various air quality models (CALPUFF, CHIMERE⁴, CMAQ⁵) or MC2⁶ – Canadian mesoscale meteorological model couple on-line with chemistry module.

¹ <http://www.src.com/calpuff/calpuff1.htm>

² <http://www.epa.gov/scram001/aqmindex.htm>

³ <http://www.mmm.ucar.edu/mm5/>

⁴ <http://euler.lmd.polytechnique.fr/chimere/chim-body.html>

⁵ <http://www.epa.gov/asmdner1/CMAQ/CMAQscienceDoc.html>

⁶ <http://www.cmc.ec.gc.ca/rpn/modcom/>

Initial and boundary condition

Information on realistic initial concentrations of modelled pollutants near the surface and their vertical distribution are crucial, especially for short period simulations (timescale ranging from hours to day). Calculations for each grid square and each layer in the modelling domain should start with possibly realistic concentration for each modelled chemical species. Boundary conditions describing inflow of pollutants from outside of the model domain are also important for the quality of the results.

Main difficulty of determining the initial and boundary conditions is not sufficient number of measurements to reproduce the three dimensional chemical fields, thus these data must be estimated from a sparse network of ambient measurements. In absence of such measurements several assumptions can be made e.g.:

- constant values of the concentration for selected pollutant;
- pre-defined vertical profile and horizontal homogeneity;
- results from the larger scale model:
 - o climatological fields (e.g. monthly average);
 - o time-varying 3D concentration fields.

The best and the most accurate solution is to use results for larger scale models. Widely used method is nesting, also called cascade mode. The idea of such approach is to provide the initial and boundary information for the domain of interest from model simulations for larger scale and coarser resolution. Depending on the methodology the same model can be used for all simulations (self-nesting) or different models can be combined. Fig. 4.1 presents the idea of self-nesting with MC2-AQ model, used in ESCOMPTE Modelling Exercise project.

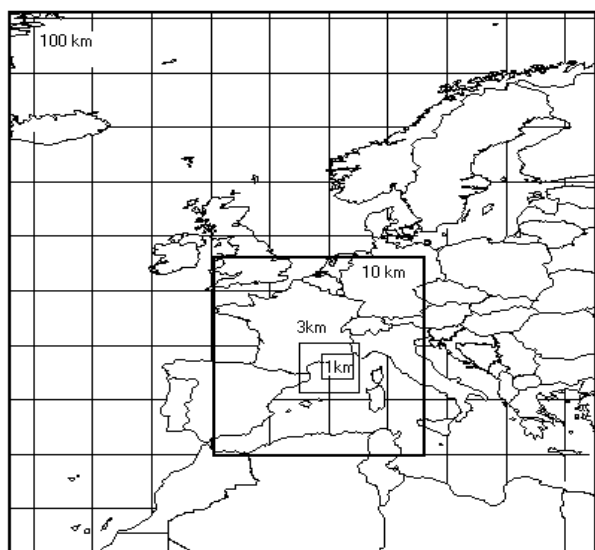


Figure 4.1. Nesting strategy for ESCOMPTE experiment 100 km → 10 km → 3 km / 1 km (example of application of MC2-AQ model, calculation performed by Warsaw University of Technology).

Primary pollutants' emission

Emission inventories are an important part of the model input. Realistic estimation of primary pollutants emission fluxes is crucial for accuracy of air quality modelling.

Emissions may originate from anthropogenic and natural (mainly biogenic) sources. For anthropogenic activities different types of emission sources are parameterised as area sources, point sources or line sources. Depending of the type of the source several parameters must be provided to the model. For point sources, required information includes stack height and diameter, temperature and velocity of exhaust gases. For the area sources, necessary information is definition of the grid for emission inventory and area per each grid square. Line sources may be

described by length of the source and traffic intensity; however, due to the model resolution in most cases such sources are included in area sources.

Species emitted from natural sources significantly contribute to the background concentration of air pollutants. If the model allows for including such information, biogenic emission of NMVOC and NO_x emission from soils should be provided. This type of emission strongly depends on meteorological parameters (solar radiation, air temperature) and in many models it is parameterised based on the landuse information. Many studies suggest that VOC emissions from plants may play a role in the formation of ozone and fine particulate matter. Unfortunately, biogenic hydrocarbons remain a potentially large and yet not well understood source of emissions.

For primary aerosols emitted from natural and anthropogenic sources the chemical speciation and size distribution depend on the aerosol module formulation. Some assumptions are made for anthropogenic fluxes of PM₁₀ and PM_{2.5} which are usually distributed among sulphates and nitrates. Natural emission of aerosols may include black carbon from biomass burning, sea salt and desert dust.

Processing of the emission inventories for the model input may cause difficulties. Main problems are connected with combining the information from different sources (Fig. 4.2) and horizontal interpolation with the respect to mass conservation. Another issue is vertical distribution of emission flux which may significantly influence modelled surface concentration. Also, speciation of NMVOC for selected hydrocarbon groups, which are specific for the chemical mechanism of the air quality model, may be the source of uncertainties of input emission data.

Official emission data reported by European countries are compiled by experts from EMEP-WSC centre and are available at EMEP web-page⁷. This data covers entire Europe with the resolution 50 km. In case of the global problems, several datasets can be used. An example may be EDGAR⁸ inventory or GEIA⁹ datasets; the resolution of this data is 1 deg (global coverage). Also, in the frame of ACCENT Network the web portal for access to emission databases¹⁰ has been developed.

For several air quality model applications the resolution of mentioned datasets is not sufficient. Preparation of emission inventories for the local scale is quite difficult and such inventories are not easily accessible.

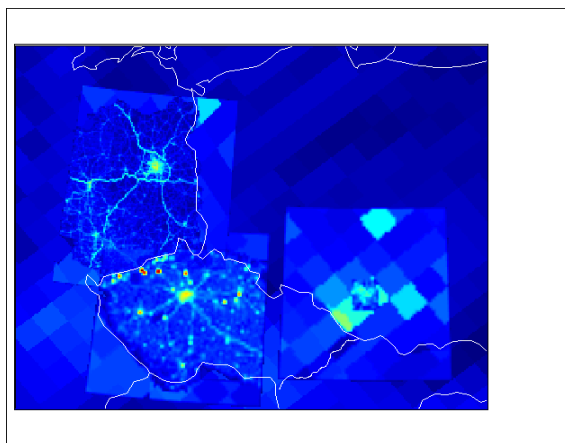


Figure 4.2. Compilation of three local inventories (Berlin, Krakow and Katowice) and EMEP inventory for CityDelta project calculations. Inconsistencies between these inventories are shown. (calculation performed by Warsaw University of Technology).

⁷ <http://webdab.emep.int/>

⁸ <http://www.mnp.nl/edgar/>

⁹ <http://geiacenter.org/>

¹⁰ <http://www.accent-network.org/>

Geophysical and landuse information

Geophysical fields are necessary for the model to characterise surface properties. The list of required fields usually includes surface roughness, topography, soil moisture, albedo, landuse classification and vegetation types. This list is specific for the model and the information should be obtained with a respect to the model domain and resolution.

5. How to start the experiment?

The strategy of the experiment should be designed before starting numerical simulations with air quality model. Clear definition of the objective of the study will help to choose an appropriate type of the model.

Necessary decisions concerning setup of the experiment are connected with:

- domain size and resolution (geographical projection, reference coordinates);
- length of the simulation (starting date, time step, number of time-steps);
- checking data requirements;
- revision of possible sources of initial data;
- processing data to produce model input.

Input data requirements are specific for the model and best possible data sets should be used to assure good quality of the modelling results.

After completing simulations the comparison of the results with available measurements should be undertaken to evaluate model performance. Information on limitations of air quality model used for calculations is necessary to assess uncertainties connected with meteorological data and emission data. Also, basic knowledge on physical and chemical processes occurring in the atmosphere ensures correct interpretation of modelled results¹¹.

¹¹recommended lecture: Jacobson M. Z., "Fundamentals of Atmospheric Modeling", Cambridge University Press, New York (1998)