Annex A.

Input information and brief model description of modeling with fine (5x5 km^2) spatial resolution for the Netherlands

A1. Preparation of meteorological and geophysical information

Meteorological data with spatial resolution 5x5 km^2 were prepared using MM5 meteorological pre-processor [*Grell at al*, 1994]. The set of MM5 parameterizations used in pre-processing of meteorological fields is described in [*Travnikov and Ilyin*, 2005]. ECMWF analysis data (http://www.ecmwf.int/services/archive/) were used as input data for MM5. Meteorological information was verified via comparison of modeled and observed meteorological parameters. Precipitation and wind velocity are the most important measurable parameters for modeling of atmospheric transport and deposition of heavy metals. Therefore, the comparison is focused on these two parameters. Observed precipitation were derived from the European Climate Assessment & Dataset (ECA&D) [*Klein Tank et al*, 2002, *http://www.ecad.eu*]. Information on surface wind velocities was taken from NOAA archives of station daily data [*ftp://ftp.ncdc.noaa.gov/pub/data/gsod/*].

Flat terrain and relatively small extent of the Netherlands cause more or less uniform climatic conditions over the country's territory. Observed annual precipitation sums vary from 800 to 1200 mm, and the modeled ones - from 800 to 1400 mm (Fig. A1a). On average, precipitation amounts produced by MM5 are somewhat higher than the observed precipitation. As seen from map of spatial distribution of observed and modeled precipitation, MM5 tends to overestimate precipitation in the northern part of the country and underestimate – in the southern part (Fig. A1b). In order to evaluate model performance of temporal variability of precipitation, correlation coefficient was calculated for 10-day sums of modeled and observed precipitation time series for each station. For most of stations this correlation coefficient is higher than 0.5 (Fig. A1c).

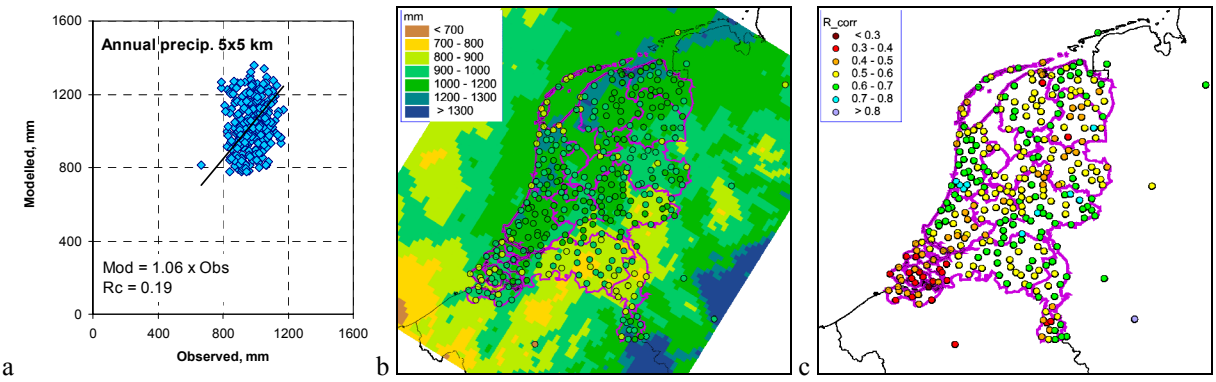

Fig. A1. *Comparison of modeled and observed annual sums of precipitation in the Netherlands in 2007: modeled vs. observed regression (a), spatial distribution of modeled and observed precipitation (b) and spatial distribution of temporal correlation coefficient (c).*

Modelled and observed annual mean 10-m wind velocities range from 3.5 to 8 m/s. The modeled values match well the observed ones: the regression coefficient is 1.01 and spatial correlation coefficient is 0.89 (Fig. A2a). Both model and observations demonstrate higher

values over sea surface and lower - over land mass (Fig. A2b). Most of daily correlation coefficients is higher than 0.8 (Fig. A2c), which indicates than temporal variability of wind velocities throughout the year is reproduced reasonably well.

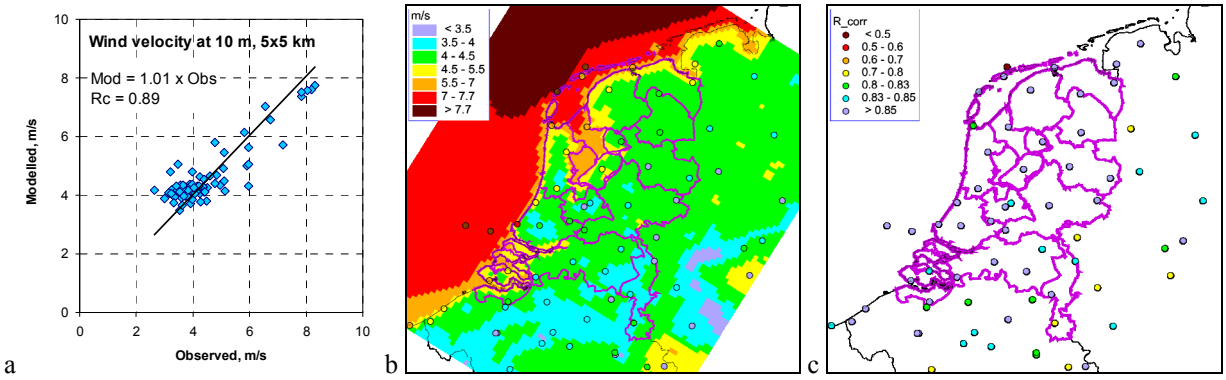

Fig. A2. *Comparison of modeled and observed annual mean wind velocities in the Netherlands in 2007: modeled vs. observed regression (a), spatial distribution of modeled and observed wind velocities (b) and spatial distribution of temporal correlation coefficient (c).*

Input geophysical information prepared for modelling with the resolution 5×5 km^2 includes data on land-cover and concentrations in soils. Land cover is important factor strongly affecting modelled dry deposition velocities. Besides, ecosystem-dependent deposition data are needed for evaluation of critical loads exceedances. Land cover map with fine spatial resolution is generated on the base of MODIS satellite information [*www-modis.bu.edu/landcover/*].

Concentrations of lead in soils are needed to simulate wind re-suspension fluxes, which can contribute significantly to total pollution levels. For example, in 2011 its contribution to deposition exceeded 50% in most countries of the EMEP region. Measured concentrations of lead in background topsoil were interpolated by Krigging method to grid with 5x5 km^2 resolution. Topsoil observations of heavy metal concentrations are derived from the European-wide project FOREGS [*www.gtk.fi/publ/foregsatlas/*]. It is important to stress that the same wind re-suspension scheme was applied both for modeling over the EMEP domain with resolution 50x50 km^2 and for high-resolution modeling for the Netherlands.

A2. Brief model description

The EMEP/MSC-E regional model of heavy metals airborne pollution (MSCE-HM) is a three-dimensional Eulerian type chemical transport model driven by off-line meteorological data. The model considers heavy metal emissions from anthropogenic and natural sources, transport in the atmosphere, chemical transformations (of mercury only) both in gaseous and aqueous phases, and deposition to the surface (Fig. A3). Operational version of the model applied for the EMEP tasks has spatial resolution of 50x50 km^2. For country-specific problems the model was adapted for calculations with fine resolution (5x5

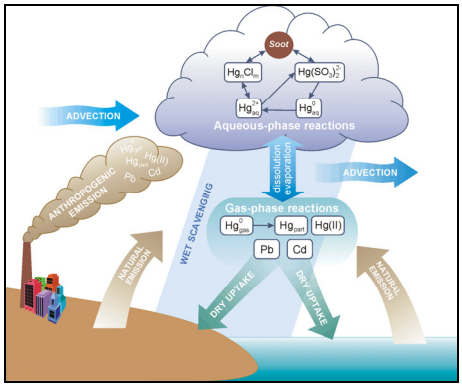

Fig. A3. *The model scheme of heavy metal behaviour in the atmosphere*

km^2) over limited domain. The vertical structure of the model is formulated in the sigma-pressure coordinate system. The model domain consists of 15 irregular sigma-layers and has a top at 100 hPa. Detailed description of the model is available in [*Travnikov and Ilyn*, 2005].

MSCE-HM model is driven by off-line meteorological data pre-processed by MM5 – Fifth Generation Penn State/NCAR Mesoscale Model [*Grell et al*., 1995]. The pre-processor utilizes the NCAR/NCEP re-analysis or ECMWF data as the input information and provides 6-hour weather prediction data along with estimates of the atmospheric boundary layer parameters with the same spatial resolution as that of the transport model.

Wind re-suspension of particle-bound heavy metals (like lead and cadmium) from soil and seawater appears to be important process affecting ambient concentration and deposition of these pollutants, particularly, in areas with low direct anthropogenic emissions. Parameterization of heavy metal wind re-suspension is included into the MSCE-HM model [*Gusev et al*., 2006; *Ilyin et al*., 2007]. The dust suspension is estimated for non-vegetated surfaces (deserts and bare soils, agricultural soils during the cultivation period, and urban areas). Generation of sea-salt and wind suspension of heavy metals from the sea surface is also considered based on the empirical Gong-Monahan parameterization [*Gong*, 2003].