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Transformations of long-living and short-living gaseous pollutants in the atmosphere of urban regions

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The research was devoted to the problem of estimation of chemical transformations of source species and atmospheric species in high-polluted areas. Box Air Quality Model (BAQM offline) was developed to estimate degree of influence of different species on atmospheric processes by analysis of chemical transformation and consequently lifetimes of these species, i.e. how long a representative molecule of the substance will stay in the atmosphere before it is chemically removed.

Preliminary study of chemical mechanisms of Global and Regional weather forecast models with chemical branch (Enviro-HIRLAM, WRF, ALADIN, ECMWF GEMS) helped to develop a universal chemical mechanism for BAQM. The new mechanism describes chemical reaction pathways for the troposphere and lower stratosphere and can be implemented at regional and global scales. The mechanism was developed using lumping technique on the basis of RACM mechanism. Aggregation of primary species into lumped species is based on their reactivities and emission rates. The different chemical solvents were used to simulate change of production and destruction.

As initial conditions BAQM considers both biogenic and anthropogenic emissions.

Lifetime calculations show that "long-living" gases demand special attention since make the greatest impact on global atmospheric processes. Such species well mix in the atmosphere and can transport for long distances from the source of emissions. “Short-living” species can affect regional processes especially in the urban polluted areas where concentration of polluted species is high. So, in such regions (large cities, industrial areas, megacities) there are high concentrations of O3, NOx, but air quality depends on distribution of these concentrations in observing region.

According to the simulations we define “long-living” species: SO2, N2, CH4, CO, H2, H2O (above 70hPa), H2O2, HCl and “short-living” species: O3, O(3P), O(1D), H2, HNO3, OH, HO2, CH3, CH3O2, CH3OOH, N, NO, NO2, NO3, Cl, Br2, BrO

Some gases such as NOx can be short-living and long-living simultaneously. Its behavior depends on different atmospheric conditions, concentrations of other gases such as OH and O3, time of the day or model domain. It should be taken into account at chemical modeling to define which species will dominate in horizontal or vertical transport.

The BAQM model confirms strong dependence of O3 on HOx. It is happened because each O3 molecule crossing the tropopause can yield at most two OH molecules in the troposphere. However, the concentrations and lifetimes depend on period of the day also. There isn’t any production or loss due to photolysis reactions at night, but at daytime the photolysis plays an important role.

The larger hydrocarbons have smaller global sources than CH4 and are therefore less important than CH4 for global tropospheric chemistry. They are however critical for rapid production of O3 in polluted regions, and play also an important role in the long-range transport of NOx.

Hence, chemical feedbacks are very important mechanisms in the atmosphere of urban areas. Since the amount of chemically active species in the atmosphere increase due to emissions from the surface layer the conditions for chemical transformations in the upper troposphere change. Consequently, the emissions of chemically active species from polluted surface areas to the atmosphere increase (positive feedback) or the emission of chemically active species to the atmosphere decrease (negative feedback).

Box Air Quality Model can be coupled with regional or global atmospheric models as a chemical module.