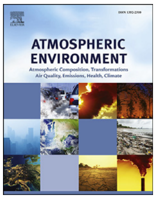
Atmospheric Environment 128 (2016) 216–226

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv



Analytical dispersion model for the chain of primary and secondary air pollutants released from point source

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HIGHLIGHTS

- We propose the model for primary and secondary pollutant dispersion in the atmosphere.
- Analytical model takes into account processes of pollutant transformation and removal.
- Used diffusivity coefficients K_i are expressed through the dispersion parameters σ_i.
- We provide analytical expressions of solution for pollutant chain dispersion.

ARTICLE INFO

Article history:
Received 11 September 2015
Received in revised form
18 November 2015
Accepted 7 December 2015
Available online 12 December 2015

Keywords:
Atmospheric dispersion
Analytical model
Point source
Secondary pollutant
Contaminant transport
Gaussian plume solution

ABSTRACT

An analytical model for dispersion of air pollutants released from a point source forming a secondary pollutant (e.g. chemical transformation or parent-daughter radionuclide chain) is formulated considering the constant wind speed and eddy diffusivities as an explicit function of downwind distance from the source in Cauchy (reflection-deposition type) boundary conditions. The dispersion of pollutants has been investigated by using the Gaussian plume dispersion parameters σ_y and σ_z instead of the diffusivity parameters K_y and K_z. For primary pollutant it was proposed to use the derived dry deposition factor instead of the source depletion alternative. An analytical solution for steady-state two-dimensional pollutant transport in the atmosphere is presented. Derived formulas include dependency from effective release height, gravitational and dry deposition velocities of primary and secondary pollutants, advection, surface roughness length and empirical dispersion parameters σ_y and σ_z. Demonstration of analytical solution application is provided by calculation of ^{135}Xe and ^{135}C air activity concentrations and the applicability of the model for the solution of atmospheric pollution transport problems.

1. Introduction

Dispersion of toxic or radioactive pollutants is a sensitive issue, especially if it results in a long term contamination. The disasters of Chernobyl and Fukushima left radioactive traces in our environment for a long time after a relatively short burst of radioactivity. Other source of contamination is routine discharges from chemical, heavy industry factories in industrial areas. Discharge to the atmosphere and transport of chemical or radioactive materials causes their dispersion and fallout on big distances from the industrial sites in case the deposition velocities are low (see e.g. Kalinowski and Pistner, 2006; Baklanov and Aloyan, 2002). Radioactive pollutants can be produced by nuclear explosions, nuclear events, war explosives, and natural processes such as the radioactive decay of radon. A special case is the dissemination of the chain of primary and secondary pollutant. The secondary pollutant forms in the atmosphere through chemical and photochemical reactions from the primary pollutants, or can be the decay products of primary (parent) pollutants which are radioactive. The transformation from primary pollutant to the secondary one can change the chemical and physical properties considerably thus influencing the final process of the dissemination and deposition on the ground surface (e.g., Delmore et al., 2011; Zhang et al., 2002). The problem of secondary pollutant needs adequate formulation and analysis due to practical reasons when secondary pollutants have longer half-life periods and may be more hazardous and more toxic than primary pollutants (see e.g. Lakshminarayanachari et al., 2013).

In this paper the proposed mathematical model considers secondary pollutant which might be generated from the primary pollutant due to radioactive decay as well as due to chemical

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http://dx.doi.org/10.1016/j.atmosenv.2015.12.019
1352-2310/

reactions and their removal by means of settling. Application of this model allows predicting pollutant air concentrations due to routine releases as well as due to accidental discharges of radioactive or toxic materials. In particular, we analyze the process of the transport of the primary and secondary pollutant in case of their discharge from elevated discharge point to the atmosphere. The purpose of this work is to develop an analytical plume model for the atmospheric dispersion of primary and secondary pollutants which are coupled by the first order transformation process. This can be chemical or photochemical reaction, or radioactive decay parameterized by a corresponding reaction (or decay) rate. In particular, the presented model treats dry deposition in the manner proposed by Calder (1961) where both the gravitational settling flux and the ground deposition flux are treated as the first order processes involving local air concentrations of pollutants. On this basis and the assumptions of the Gaussian plume model with dispersion parameters, the atmospheric transport equations are solved for primary and secondary pollutants and analytic expressions are obtained for the air concentrations of primary and secondary pollutants. Analytical solution expressions involve Gaussian plume dispersion parameters which are more advantageous than K_y and K_z due to their availability for various atmospheric conditions and wide practical use. Moreover, analytical solutions provide possibility for rapid evaluation of pollutant air concentrations, which can be preferable in case of the need of fast forecast of pollutant dispersion, e.g. forecast of pollutant concentration in case of an accident. On the other hand, analytical solutions can be used for the verification of corresponding numerical solutions, because the latter convergence is not always established and specialized procedures are being applied (see e.g., Lakshminarayanachari et al., 2013).

It is shown by De Visscher (2014) that the use of Chamberlain techniques (Chamberlain, 1953) to manage deposition allows assessment of primary or secondary pollutant air concentrations by Gaussian plum model which does not always lead to the desired result in terms of accuracy. Therefore in addition to this method other more complex methods were developed able to assess both dry deposition and gravitational settling (Horst, 1977; Dorm and Horst, 1985; Alex De Visscher, 2014). The methods are used to describe the primary and secondary pollution as well as the gravitational settling where the flux is proportional to the pollutant concentration in air as it was proposed by Calder (1961). In this paper we implemented these assumptions additionally to the ones of the main Gaussian plum model and derived analytical expressions of air pollution concentrations for secondary pollutant. From physical point of view these assumptions are more realistic than that obtained using the source depletion model, preserving as much as possible the concept of the Gauss plum model.

2. Rationale of the model development

In this paper we analyze the case of the dissemination of primary and secondary pollutant chain discharged from the elevated point to the atmosphere. The problem is that the transformation from primary to secondary pollutant takes place during the dispersion in the air. This transformation can be chemical or photochemical reactions, radioactive decay, etc. In general the chemical and physical properties of the primary and secondary pollutant can be very different, i.e., this has to be taken into account explicitly in the mathematical modeling.

A good example of change of the properties of primary and secondary pollutant is the case of the dissemination of discharged radioactive noble gasses from operating nuclear power plants (NPP's). A number of radioactive isotopes are generated as the decay products in the nuclear fuel and in the presence of fuel cladding defects they are released to the coolant and finally can escape to the atmosphere under normal operation of NPP's: ^{85}Kr, ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{133}Xe, ^{133m}Xe, ^{135}Xe, ^{138}Xe. An overview of radioxenon emissions is provided for global NPP fleet by Kalinowski and Tuma (2009). Moreover, the presence of many radioactive xenon isotopes provides a possibility to distinguish between gaseous releases of LWR and nuclear explosion tests on the basis of the radioisotope activity ratio (Kalinowski and Pistner, 2006). The special case is the ^{135}Xe isotope (half-life 9.2 h) which decays to the long-lived ^{135}Cs (half-life 2.3 × 10^{6} years). The half-life of ^{135}Xe is long enough to escape from the fuel through several barriers of NPP to the atmosphere and to travel a relative big distances due to its chemical inactivity until its decay to ^{135}Cs (Remeikis et al., 2012). Once ^{135}Cs is formed, it is rapidly attached to the aerosol particles, which will effectively settle on the ground surface, plants or can be washed out by rain. Therefore transformation from primary to secondary pollutant is a turning point in the process of the pollutant migration from the discharge point to the final fallout location. Secondary pollutant problems have been studied by various authors. These models range from the most complete but computationally most sophisticated and time extensive to relatively simple Gaussian and box models (e.g., Daly and Zannetti., 2007; Baklanov et al., 2009; Visscher, 2014). The three dimensional Computational Fluid Dynamics (CFD) models can be attributed to the class of the most sophisticated ones (e.g. ARIAL, MISKAM), based on the transport theory of mass, momentum, energy and species. Another class of models are Lagrangian/Eulerian type ones (e.g. AUSTAL2000, CALGRID, CIT, DRAIS, MARS, UAM, TAPM, ARIA, DEHM, SMAQ, CAMx). The Lagrangian model follows the trajectory of the box/particle with initial concentration as it moves downwind. It incorporates changes in concentration due to mean fluid velocity, turbulence of the wind components, molecular diffusion and chemical transformations. Similar Eulerian advection and dispersion models are based on a grid that is fixed in space. In each grid point the advection and dispersion of pollutants is calculated proportional to local concentration gradients by using turbulent version of Fick's law (Uma et al., 1991). Gaussian models assume Gaussian type distribution of pollutant concentration profile and width of the plume which is determined by distant dependent σ_y and σ_z, which are defined by atmosphere stability classes (Pasquill, 1961; Gifford, 1976), or additionally takes into account plume spread dependence on the local wind speed and turbulence and thus depends on plume height (Carruthers et al., 1999), but few of the Gaussian models are able to deal with secondary pollutants (e.g. plume model AERMOD and puff dispersion models CALPUFF, RIMPUFF or combined model ADMS).

Numerical methods are used to solve the transport and transformation equations in CFD and Lagrangian/Eulerian models. This usually requires a high computational effort and sometimes it is impossible to study the contaminant dispersion in complex and large domains, especially in the surface layer where a large range of length scales are involved. Moreover, application of the numerical methods requires careful reliability study by considering consistency, stability and convergence (Lakshminarayanachari et al., 2013). In contrast to complex numerical models the Gaussian model can be implemented as an analytical solution or by using relatively simple numerical calculation techniques.

Analytical solutions of the steady state three-dimensional atmospheric diffusion equation from point source are presented, e.g., by Alam and Seinfeld (1981). The solution permits prediction of the three-dimensional concentration distribution of primary pollutant converted to secondary product by atmospheric chemical reactions, and both are removed by wet and dry deposition. Effects of removal by chemical transformation, washout and etc. during the dispersion