A REVIEW ON SHORT-RANGE DISPERSION MODELLING OF GASEOUS POLLUTANTS RELEASED FROM STATIONARY SOURCES

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The reliance on solid fuels for heat processing and power generation with low efficiency and inadequately control of gaseous pollutants is strongly related to health impacts and environmental pollution. Atmospheric dispersion modelling is an essential tool to assess the potential impacts of related stationary sources on local air quality. Advanced Gaussian plume models provide an up-to-date assemble of algorithms to estimate contaminant concentrations distributed at a variety of heights and distances. This review presented a comparative evaluation on ADMS and AERMOD performance in different validation scenarios considering SO_2 emissions and the NO_x chemistry scheme. Terrain, stack height and plume rise influence on emissions, along with modelling uncertainties and limitations were also discussed. Contour plots of maximum daily values and annual averages confirmed a remarkable similarity in patterns within simulations. By this approach, the study extended recent practical information and recommended a complementary instrument for the improvement of the reference model SYMOS´97 implemented in the Czech Republic.

Keywords: power plant, air dispersion modelling, Gaussian plume model, nitrogen oxides

Received 18. 05. 2020, accepted 15. 06. 2020

1. Introduction

Energy security plays a key role in every modern economy today. Despite a rapid development in renewable energy diversification, coal, oil and natural gas are still the world's primary energy options, with coal-fired power plants being responsible for 38% of the global heat and power generation [1]. Economic development and growing demands of energy have increased the utilization of coal and fossil carbon-derivative solid fuels given their relatively low extraction cost and price stability in the market [2].

The most common and simplest utilization of mineral fuels and solid biofuels is through direct processing in thermochemical conversion systems for heat generation and electricity production, or as feedstocks to produce coke and coal gas [3]. In the conventional power plants, co-firing thermal stations or municipal solid waste incinerations, solid fuels are burnt at high temperatures. Low conversion efficiency and inadequately emission control potentially release a considerable amount of flue gases, and so contributing to the atmospheric pollution phenomena.

From this perspective, the significant dependence on solid fuels for energy production (amounting to 50% of the electricity generated) in the Czech Republic has been directly related to air quality issues and detriment of public health [4]. Carbon Majors Report in 2017 identified that coal related industries in the country are among the world's 100 biggest environmental polluters [5]. Moreover, the energy sector contributes the most to sulfur oxides, nitrogen oxides, heavy metals and polychlorinated compounds emissions [6].

Specifically, energy production, distribution and use are responsible for almost 45% of NO_x emissions [6]. Besides contributing to acid deposition and eutrophication of soil and water along with sulfur oxides; this group of nitrogen oxides is associated with the formation of ozone and particulate matter; and it is related to adverse human health effects (e.g. liver, lung, spleen diseases) [7]. Namely, secondary pollutants such as nitrates, can account for 95% of total health impacts from coal used for power generation in the Czech Republic [8].

Dispersion modelling is introduced as a useful methodology to assess the potential impact of this sector on air quality. It consists in the development of mathematical expressions to describe the planetary boundary layer (PBL), the flow field and displacement of the plume of contaminants in the atmosphere, taking into account the chemical and physical processes that govern its behavior. Such modelling tools have been broadly used to predict concentrations of hazardous compounds in the environment and provide relevant information for specific sectors and decision makers [9].

In practice, Gaussian plume models are still the most extensively used tools for regulatory purposes because of their robust model setup and easy implementation. They are based on the Gaussian distribution under steady state conditions with two dispersion parameters, σ_y and σ_z, which determine the width of the plume in the horizontal and vertical directions [10]. Unlike Lagrangian or Eulerian models, instead of solving differential equations, Gaussian models stand out by performing almost immediate calculation, since they only compute a single formula for each receptor point [11].

Hence, computational cost mainly depends on meteorological data pre-processing and turbulence parametrization, allowing a near real-time decision support [11]. Nonetheless, plume dispersion models assume that meteorological conditions remain constant between the time of emission and arrival at the receptor, which limits the range of application.

Furthermore, there are constraints of plume models predicting secondary organic aerosol formation and estimating concentrations under low wind speeds or at sites close to the source [10]. The latter is one of the most worrying limitations given the importance of predicting concentrations in such dangerous conditions, i.e. stable boundary layer or low-level inversions [11].

For this reason, the performance of typical local dispersion models was revised and evaluated in this specific research. Given the importance of using a modern tool for evaluating air pollution impacts in the Czech Republic, ADMS and AERMOD were taken into consideration in order to complement the reference model SYMOS'97. In this sense, the initial aim was to address the differences between the technical capabilities of those models to simulate dispersion of gaseous pollutants from stationary point sources.

Additionally, the chemistry schemes for NO_x calculations in ADMS and AERMOD were reviewed. A hypothetical scenario was used to evaluate the impact of NO_x emissions through ADMS, AERMOD and a hybrid model that runs AERMOD with ADMS met pre-processor. This was essential to further acknowledge the differences in the meteorological and dispersion algorithms.

2. Plume dispersion modelling

2.1. ADMS

The Advanced Dispersion Modelling System (ADMS), developed by Cambridge Environmental Research Consultant (CERC), is a Gaussian-like steady state model able to simulate continuous plumes and short duration puff releases [12]. It stands out from other regulatory models due to its ability to consider radioactive decay; model the effect of wind turbines; study plume visibility, its temperature and humidity; and take into account the effect of coastal regions and offshore areas [13].

In addition, ADMS has an integrated meteorological pre-processor to calculate important boundary layer parameters and it is supplied with Mapper, a tool used to visualize, add and edit sources, buildings and output points [13]. In order to describe the distribution of pollutants, the modelling system assigns a normal Gaussian distribution in stable and neutral conditions, while distinguishing the horizontal dispersion (Gaussian) from the vertical dispersion (bi-Gaussian or skewed Gaussian) in a convective boundary layer [10].

2.2. AERMOD

The AMS/EPA Regulatory Model Improvement Committee (AERMIC) developed AERMOD in order to estimate near-field impacts from industrial sources, introducing the recent knowledge on PBL concepts and including the treatment of simple and complex terrains [10]. In this manner, it was intended to replace ISC ST3 (Industrial Source Complex Model), the US EPA preferred air dispersion model for industrial sources in simple terrain, which is based on Pasquill-Gifford stability classes [12]. Although AERMOD was built upon the same framework, it allowed to account for changes in dispersion rate with height, which enabled a non-Gaussian plume representation in convective conditions [14].

Furthermore, AERMOD upgraded the algorithms to calculate dispersion in stable, convective and urban night-time boundary layer; plume rise, buoyancy and penetration into elevated inversions; building wake effects, among others [15]. For this purpose, the modelling system contains two pre-processors: a meteorological pre-processor (AERMET) and a terrain pre-processor (AERMAP). AERMET estimates hourly boundary layer parameters, for instance friction velocity, Monin-Obukhov length and mixing height; and prepares vertical profiles of wind, turbulence and temperature [16]. On the other hand, AERMAP facilitates the characterization of the terrain with hill height scales and generates receptor grids for the executable runs [16].

2.3. SYMOS'97

The Czech Hydrometeorological Institute (CHMI) developed the *Systém modelování stacionárních zdrojů* (SYMOS) as a Gaussian reference model for calculations of non-reactive releases from single or multiple sources. The boundary layer is described through the specific stability scheme proposed by Bubník and Koldovský, based on routine observations from synoptic meteorological stations in the Czech Republic [17]. It includes five stability classes: three of these describe stable stratification, ranging from strong to weak inversions, and the rest cover neutral and convective conditions [18]. Nevertheless, it has some limitations, e.g. restrictions for calculations under inversed layers in complex terrain, sources below roofs of buildings, urban modelling. The main characteristics of these models are summarized in Table 1, describing technical features which are important for emissions from stationary sources.

3. Performance on stationary sources

In this aim, the validation of dispersion models is normally carried out injecting tracer gases (for example sulfur hexafluoride, SF6) into the buoyant combustion gas or measuring emitted SO_2 concentrations released from the stack of thermal power plants [12].

However, SYMOS belongs to a group of models which are yet to be extensively validated against measured pollutant concentrations near point sources [23]. From this perspective, ADMS and AERMOD were analyzed in-depth and discussed.

3.1. Flat terrain

A frequently indicated study on a flat rural terrain involved short-term intensive measurements (1-h averages) in Kincaid (Illinois) with a highly buoyant released from a tall stack emitting SF6.

Table 1. Summary of modelling features relevant to atmospheric dispersion of stationary emissions

Features	ADMS	AERMOD	SYMOS'97
Application	Applicable up to 60 km, provides information up to 100 km	Up to 50 km from sources	Recommended up to 50 km from sources
Source types	Point, line, area, volume, grid, jet sources	Point, line, area and volume sources	Point, line and area sources
Meteorological pre-processor	Built-in module	AERMET	CALMET/ALADIN
Input of vertical profiles of meteorological data	Yes (optional)	Yes (required)	No
Boundary layer structure	Boundary layer depth and Monin-Obukhov length	Boundary layer depth and Monin-Obukhov length	Bubník & Koldovský stability classes
Plume rise	Advanced integral model	Briggs and Weil empirical expressions	Modified Briggs expressions
Buildings downwash	Two region model extending to downwind distance of 60 building heights. Based on flow model with near and main building wakes	PRIME module with BPIP pre-processor. Two region model with influence of building decreasing exponentially	No
Complex terrain	Based on calculation of flow field and turbulence field by FLOWSTAR model	Interpolation between plume displaced by terrain height and plume impaction	Combination of lower and upper concentrations estimated using a weighting factor
Deposition (wet and dry)	User-defined deposition velocity or resistance model (single value over domain)	User-defined resistance model based on land cover type (varies by wind direction/season)	Only for coarse particles through sedimentation velocity
Chemistry	Generic reaction scheme for NOx, amine chemistry, parameterised sulphate chemistry	Ozone limiting or plume volume molar ratio models	Decay term and basic scheme for NO-NO2 (simple parametrisation using residence time)

Note. Data for ADMS and AERMOD [19] and [20], data for SYMOS'97 [21]

Perry et al. [22] reported that AERMOD had the tendency to underpredict higher concentrations, as well as Hanna et al. [12], which obtained underpredictions for both AERMOD and ADMS (52% and 34%, respectively).

Nevertheless, while AERMOD accurately predicted the location of the observed maximum, ADMS still showed better performance with an average bias of 3% and a higher fraction of predictions within a factor of two, with 59% against 29% [12].

Moreover, a long-term study was done in Kincaid with highly buoyant SO_2 emissions. AERMOD captured the upper end of the distribution considerably well for the 3-h and 24-h averaging periods, but the annual average remained particularly underpredicted [22].

In comparison, AERMOD and ADMS have been further assessed in a flat urban/commercial setting in Indianapolis power plant, where they showed minor overpredictions (13% and 7%, respectively) [22]. This study suggested that AERMOD tends to underpredict on the closest arcs and overpredict on the furthest arcs, whereas ADMS did the opposite [24].

Additionally, a hybrid version of ADMS and AERMOD (i.e. AERMOD was run using the ADMS met pre-processor) has been compared against ADMS to analyze typical stationary sources of a variety of heights (near ground, 50 m and 199 m); with and without plume rise; for short-term (convective, neutral and stable conditions) and long-term meteorology using Clifty Creek (Indiana) field experiment data, in order to account only for the differences caused by dispersion calculation [25].

The simulated outcomes identified that short-term maximum concentrations were similar especially in cases without plume rise, showing even a comparable plume shape. On the other hand, for long-term meteorology, differences were noticeable in low-level stacks [25].

3.2. Complex terrain

A notable short-term study in mountainous terrain was performed in Tracy power plant, where stable conditions were predominant [22]. The distribution of AERMOD significantly matched throughout the data range, and along with ADMS, results were nearly unbiased at the top end of the concentration distribution, with a slight overprediction [14, 26].

Moreover, AERMOD was tested against two long-term, complex terrain studies with tall stacks and highly buoyant SO_2 emissions (Lovett power plant and Martins Creek steam electric station). The model slightly overpredicted concentrations for 3-h and daily averages, while annual averages were generally underpredicted due to a drop-off in the distribution of low concentrations [14].

Likewise, Hanna et al. [12] proved AERMOD was within 1% of the highest concentrations for Lovett data, while ADMS underpredicted the peak concentration by 40%, although an earlier version of the model was used [14]. Meanwhile, ADMS consistently underpredicted concentrations for all averaging times in the Martins Creek study [27].

In another research work, Carruthers et al. [28] compared ADMS and AERMOD (with ADMS met pre-processor) to assess their complex terrain algorithms, ignoring plume rise. One of the cases evaluated was Clifty Creek power station where emission sources are located above a deep valley within a relatively flat plateau area.

The baseline was a comparison of both models assuming a flat terrain, in which remarkably similar patterns were displayed. However, calculations with complex terrains showed that ADMS predicts a higher impact in Clifty Creek case study (35% increase in annual average), yet the influence in AERMOD was relatively small with a subtle decrease [28].

3.3. Buildings downwash

Perry et al. [22] studied the performance of AERMOD against two datasets that emphasize on near-field concentrations resulting from building wake effects (Bowline power plant and Lee power plant). In the first case, while the ratios of modelled to observed robust highest concentrations for AERMOD were 1.14 (3-h), 1.43 (24-h) and 1.5 (annual) [22], ADMS underpredicted consistently with ratios of 0.95 (3-h), 0.62 (24-h) and 0.23 (annual) [29].

Despite this, AERMOD has a significant poor estimation of the lower end of the concentration distribution below 20 $\mu g/m^3$ [30]. If the whole range of modelled values were to be analysed, results simulated by ADMS were much better with 1.03 (3-h), 1.04 (24-h) and 0.4 (annual) ratios of maximum modelled concentrations to observations [29].

Similarly, in Lee power plant, a wind tunnel experiment was established, in which AERMOD overpredicted by more than a factor of two, considering stable conditions [22], and underestimated by half the highest concentrations in neutral conditions [30]. On the other hand, ADMS was tested against the same dataset under neutral conditions, for which better results were obtained with a slight underprediction [31].

4. Nitrogen oxides chemistry scheme

When NO_x gases (NO and NO_2) are emitted from a combustion process, the NO_2/NO_x in-stack ratios are between 5-20%, while downstream of the sources, this ratio can be up to 50-90% depending on solar intensity and ambient pollution levels [32].

Consequently, it is important for atmospheric dispersion models to allow for chemical reactions to predict NO_2 concentrations at the receptors. For short time scales, the most important chemical reactions can be described by the following system:

$$NO + O_3 \rightarrow NO_2 + O_2$$

$$NO_2 + h\nu \rightarrow NO + O_3$$

where $h\nu$ implies a photon of solar radiation at the specific wavelength $\lambda < 420$ nm.

4.1. ADMS method

The current methodology used by ADMS considers both chemical equations with reaction coefficients taken from the Generic Reaction Set chemistry scheme, a semi-empirical photochemical model suggested by CSIRO [33, 34]. First, NO_x and primary NO_2 ensemble plume concentrations at the receptor are determined from the dispersion routines. Then, ADMS makes use of the instantaneous plume spread to determine the O_3 available for chemical reactions with a cross-sectional area entrainment method [35]. Finally, a fifth order Runge-Kutta scheme is used to solve the system of chemical equations and calculate final concentrations at each receptor [32].

4.2. OLM and PVMRM2

AERMOD includes two chemistry methods to predict NO_2 concentrations, the ozone limiting method (OLM) and the plume volume molar ratio method (PVMRM2). In OLM, primary NO and NO_2 concentrations due to each emission source are determined by the dispersion model. Next, secondary NO_2 at the receptor is determined from the oxidation of NO by O_3, assuming an infinite rate constant. NO is considered to be the sum of all plumes contributing to the receptor and O_3 is assumed equal to background concentration [32]. Likewise, PVMRM2 determines NO_2 from NO oxidation assuming an infinite rate constant but the amount of O_3 available for reaction is determined from the dilution of the instantaneous plume using a volume-based entrainment method [35].

4.3. Case study

With the aim of identifying the contrast between the meteorological and dispersion algorithms, ADMS 5.2.2, AERMOD (using ADMS met pre-processor) and BREEZE AERMOD v.19191 (using AERMET), were

used to simulate a hypothetical buoyant source of NO_x at a emission rate of 10 g/s from a stack with a physical height of 45 m. A set of hourly sample data at UTM 12N was employed. For the first two modelling systems, ADMS met pre-processor calculated the vertical profiles, while for the third one, measurements were used as input for AERMET. The wind rose has predominant winds from the east and southwest, as shown in Figure 1.

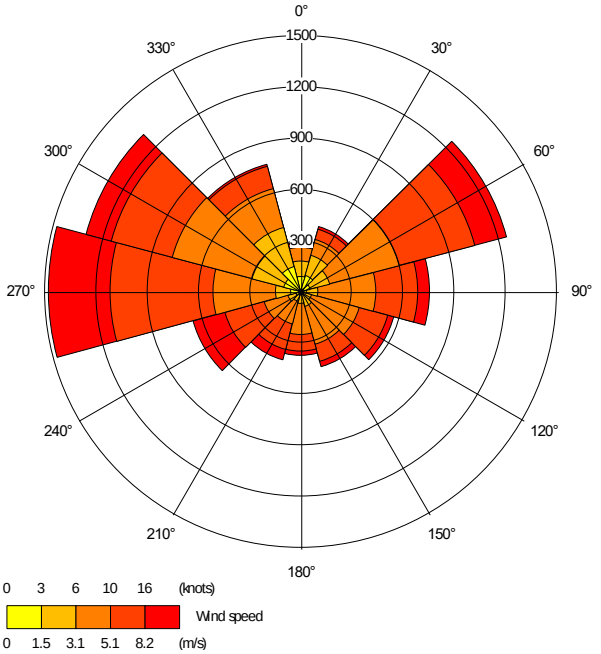

Figure 1. Wind rose from sample data

The simulated results of 1-h, 24-h and annual averaged concentrations are summarized in Table 2. The obtained maximum values for these averaging times differ greatly, especially the maximum hourly average. ADMS predicts a concentration which was almost three times the one estimated by AERMOD dispersion modules. Also, while the maximum for 24-h and annual averages occur on the same location for ADMS and the hybrid model, 1-h averaged maximum was found further to the east using AERMOD dispersion module. Moreover, it can be seen BREEZE AERMOD maximum predictions tend to be located further away from the source in the horizontal direction (daily and annual averages).

In addition, Figure 2 illustrates the distribution of NO_x concentrations on a grid of 3 km x 3 km for the daily and annual averages. The asterisks on the contour plots mark the maximum values, which are similar with the three models. However, ADMS predicts a higher impact on the source surroundings, reaching higher levels of concentration.

On the other hand, for further distances, there was a substantial agreement between the three models. Also, annual average contours show a comparable shape, especially while using the same meteorological pre-processor.

5. Discussion

5.1. Influence of terrain and building

ADMS and AERMOD showed that they can predict concentrations of gaseous pollutants with sufficient accuracy in most of the simulated scenarios. Perry et al. [22] indicated that, AERMOD had a greater success reproducing concentrations for buoyant, tall stack releases in moderate to complex topography (Lovett, Martins Creek, Tracy), compared to studies in flat and rural settings.

Nevertheless, regardless of the terrain, AERMOD estimations tended to be substantially under the actual observations for the distribution of lower concentrations. There are two reasons for this: in short-term studies, such as Kincaid (SF6), this has been explained because maximum concentrations from elevated plumes were normally associated with convective conditions, while the drop-off was a result of stable conditions [22]. Long-term studies further highlighted the challenges in reproducing lower concentrations in stable conditions, given the noticeable underpredictions for annual averages [22].

The study of Carruthers et al. [25], comparing ADMS against AERMOD (ADMS-met), also revealed that plume rise was a key driver for differences between models. In contrast with AERMOD, which calculates the plume rise through Briggs empirical expressions, ADMS employs Runge-Kutta numerical methods to make an estimation based on momentum fluxes and temperature differences between the plume and the surrounding air [10]. In addition, there is a disagreement in the obtained values from low-level stacks, suspected to be a consequence of near-source plume rise and upwind diffusion predicted by AERMOD, whereas ADMS only predicts such phenomenon in the case of calm conditions [25].

Table 2. Maximum 1-h, 24-h and annual averaged concentrations of NO_x

	ADMS		AERMOD (ADMS met)		BREEZE AERMOD	
Averaging period	Position (m)	Maximum ($\mu g/m^3$)	Position (m)	Maximum ($\mu g/m^3$)	Position (m)	Maximum ($\mu g/m^3$)
1-h	(100,0)	353.62	(200,0)	122.64	(100,-200)	111.22
24-h	(-300,-100)	41.34	(-300,-100)	32.54	(-400,-100)	31.07
Annual	(400,-100)	5.11	(400,-100)	4.33	(600,-100)	2.80

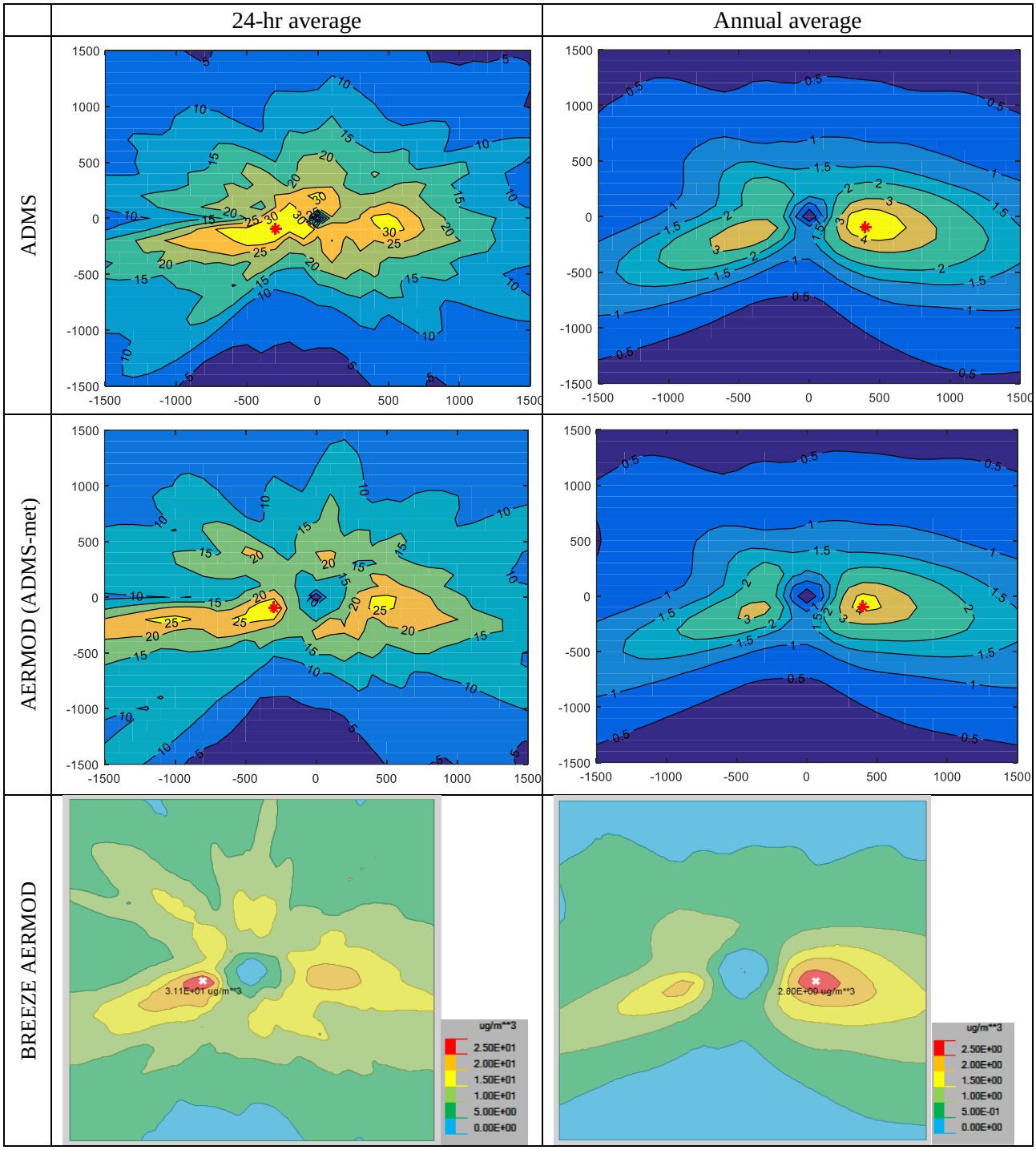

Figure 2. Contour plots of daily and annual NO_x averages using ADMS, AERMOD (with ADMS met pre-processor) and BREEZE AERMOD for long-term data.

Furthermore, the complex terrain comparison using the same methodology (i.e. running both models with ADMS met pre-processor) exhibits significant differences. This is a consequence of FLOWSTAR model predicting convergence of the main streamlines as the airflow from the prevailing direction flows out of the valley, bringing the plume closer to the surface and increasing concentrations [28]. In spite of performing reasonably well in other studies carried out in Clifty Creek [22], AERMOD's approach estimates a lower impact because much of the terrain is below the height of the stack [28].

Finally, a number of studies on building wake indicated that, there was an apparent sensitivity of dispersion to meteorology (hence to plume rise) which heavily influenced results in Lee Power Plant. Besides, Perry et al. [22] suggested the specification of the cavity extent and plume material height and spread were critical to simulate the downwash effect. Therefore, AERMOD modest overpredictions in Bowline power plant were considered as the result of a higher incidence of

downwashed plumes [22]. Conversely, ADMS had a tendency to predict lower maximum concentrations than the actual measurements. However, this is a usual feature of a model that has been developed to represent the ensemble mean i.e. a model that neglects turbulent fluctuations [29].

5.2. NO_x dispersion modelling

There are two fundamental differences between the mechanisms used by ADMS and AERMOD to model the chemistry scheme of NO_x: ADMS module includes reactions for both titration of NO by ozone as well as NO_2 photolysis, and it accounts for chemical reaction rates.

In a study carried out by the American Petroleum Institute (API) [36], the comparison between these methods using five different datasets lead to the following conclusions: OLM and ADMS generally overpredicted NO_2 concentrations, while PVMRM2 demonstrated the best mean values of NO_2. Despite this, ADMS had a reasonably higher proportion of values within a factor of two of the observations and showed the most consistent performance for the correlation NO_2/NO_x.

As a result, OLM can be considered as a screening model as it calculates an upper bound assuming NO reacts with background ozone and no photolysis occurs [32]. Meanwhile, PVMRM2 and ADMS have a better estimate of in-plume O_3, although PVMRM2 has an inconsistent behavior and has proved to surpass overestimations by OLM in some cases [36].

Notwithstanding, ADMS also shows some limitations. For instance, the reaction time is calculated as a concentration-weighted average of the travel time from each source to the receptor, instead of a continuous parameter [35]. Also, it neglects the influence of building-induced turbulence, which would result in an increase in the rate of O_3 entrainment [35].

In an attempt of assessing the performance of ADMS and AERMOD predicting the impact of NO_x emissions, which deeply influences the estimation of NO_2, it was clear that the meteorological pre-processor plays a very important role.

Considering the location of the maximum concentrations, similarity was identified between ADMS and AERMOD (ADMS-met), but values of the hybrid model resemble more those of BREEZE AERMOD, differences between the position could be attributed to the meteorological pre-processors, which comprise different algorithms to calculate the surface heat flux, boundary layer height and the mixed layer velocity scale [28].

It is worth noting that even though the hybrid model used the met pre-processor estimation of the vertical profile, the concentrations were comparable to those of BREEZE AERMOD, which highlights the ability of ADMS to calculate these variables.

Additionally, the observed patterns on the contour plots comparatively verified the results reported by Hanna et al. [24], in which AERMOD tends to underpredict in the closest arcs, while ADMS overpredicts these concentrations.

6. Conclusions

A comparative evaluation was performed within Gaussian plume simulations in different conditions in which they had been validated. ADMS and AERMOD were able to predict the upper distribution of concentrations, which is more relevant for permitting purposes and regulatory applications.

The formulation of these advanced dispersion models allows to represent the PBL accurately and capture the behaviour of pollutants under convective conditions considerably well. While SYMOS may provide reasonable results within 50 km from the source, it is still based on atmospheric stability classes, assuming that near-ground meteorological measurements represent the whole boundary layer adequately.

Furthermore, recent algorithms developed for characterizing complex terrain, buildings downwash, deposition and chemical processes outlined essential complements to the reference model in the Czech Republic, specifically for the purpose of assessment and prediction of potential impacts from stationary sources on air quality.

Above all, AERMOD tends to underpredict over flat and rural terrains and overpredict through urban and complex terrains. ADMS has a tendency to underpredict in most scenarios, although it shows a higher accuracy than AERMOD. On the other hand, by comparing the chemistry schemes it was noticeable that ADMS method is based on a more robust basis and presented consistency with actual observations. In order to model stationary sources, it is recommended to use ADMS for its advanced algorithms and proficiency at reproducing the concentration distributions.

Acknowledgements

This research was carried out under the Programme Erasmus+, Key Action 1, IMETE. The authors would express an appreciation to the European Commission for their contributions. An acknowledgment would be extended to the modelling expert group in the Czech Hydrometeorological Institute for their excellent collaboration and consultancy during the project.

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