

IDŐJÁRÁS

*Quarterly Journal of the Hungarian Meteorological Service
Vol. 121, No. 2, April – June, 2017, pp. 101–115*

Spatial and temporal pattern of pollutants dispersed in the atmosphere from the Budapest Chemical Works industrial site

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(Manuscript received in final form July 25, 2016)

Abstract— In April 2015, a serious industrial pollution gained large public interest in Budapest, the capital city of Hungary. An abandoned industrial site of the former Budapest Chemical Works company was found to contain 2000–3000 tonnes of leaking industrial waste and dangerous chemicals. As the former factory is located in a densely populated urban environment, serious public health concerns have risen. The pollution has been transported for several years from the industrial site to the neighboring areas by two ways: with groundwater transport and in the atmosphere.

This study attempts to estimate the main characteristics of atmospheric transport of pollutants originating from the Budapest Chemical Works site. The 13-year long dispersion pattern is investigated using a Gaussian dispersion model, taking into account the strong weather dependence of emission rates through deflation and evaporation. The very limited information of the amount, composition, and temporal evolution of the leaking chemicals makes it impossible to provide emission estimates; however, the spatial distribution and temporal characteristics of the pollution can be investigated.

Weather-dependent emission rate was found to be the dominant factor of the pollution, counteracted by the atmospheric stability. Largest concentrations were present in the spring and summer and during the day, while nighttime emissions were generally weak. The main direction of the dispersion was towards the south-east, however, deflating and evaporating materials showed largely different results.

Key-words: air pollution, industrial emission, Gaussian dispersion model

1. Introduction

On April 7, 2015, a short public visit was organized to the abandoned industrial site of the Budapest Chemical Works (Budapesti Vegyiművek) company in Budapest, Hungary. The visit revealed that a large amount of dangerous material was stored in leaked barrels in the open air, including carcinogenic pesticides, aromatic hydrocarbons, and isopropanol. As the company had gone bankrupt in 2007 with a confused ownership background, the responsibility has been subject of debates and a legal case. To avoid further pollution and a potential fire, authorities have decontaminated the site by January 2016 (Hevesi, 2016).

The site of the Budapest Chemical Works is located in the south-eastern part of Budapest, on a flat terrain covering an area of 0.15 km². It is surrounded by warehouses, other factories, and a railyard from the northern and western directions (Fig. 1). The Danube river flows 1.6 km to the west. To the northeast, a residential area is located approximately 1 km away from the site with a busy road laying in between. To the southern and south-eastern directions, a densely populated residential area lies. The factory and the residential area is divided by an approximately 200 m wide wooded gap.

Due to the complex business and legal background, very few information has become available on the amount and chemical composition of the waste and the pollution in the nearby areas, as well as on the temporal period of the release. The company went bankrupt in 2007, however, an environmental inspection report had already warned of serious threats around the site in 2003 (Dura, 2003). Despite the early warning on potential threats, no measurements are known until 2015, when groundwater and dust analysis was performed by independent laboratories (Greenpeace, 2015; Wessling, 2015).



Fig. 1. The area of the Budapest Chemical Works (red shape) and DDT/DDD detections in soil and dust samples (red circles) (Greenpeace, 2015; Wessling, 2015). The industrial areas to the north and west are divided by a narrow band of vegetation from residential districts to the south-southeast. Background map © OpenStreetMap contributors, CC-BY-SA.

The largest environmental concern within the industrial site is clearly the groundwater pollution. Water samples were polluted above threshold with DDT (dichlorodiphenyltrichloroethane), PAHs (polycyclic aromatic hydrocarbons), benzene, and several other components (Wessling, 2015). However, groundwater pollution is localized to a small area ranging a few hundred meters around the site, and groundwater transport is directed towards the Danube river avoiding residential areas (Dura, 2003; Wessling, 2015). Chemical composition of soil and groundwater pollution in samples taken 600–800 m from the site (Fig. 1) suggested that DDT was rather transported in the atmosphere, and not by groundwater (Wessling, 2015).

Atmospheric transport of dust and gases is a great concern for residential areas. It can cover a distance of tens of kilometres, thus affecting several thousand people. Therefore, the investigation of atmospheric dispersion patterns is of key importance. As there was no direct release to the atmosphere, the emission occurred in two ways: by deflation of the polluted dust, and by the evaporation of liquid materials. The former is primarily responsible for the DDT pollution, and the latter is more important in case of aromatic hydrocarbons. Unfortunately, there is no data on the exact amount and chemical composition of the stored materials, and there is no further chance to retrieve this information as the site has been decontaminated. In the lack of any reliable information, no emission estimate is attempted; however, a sensitivity map can be produced using an atmospheric dispersion model to estimate the spatial distribution of the pollution taking into account the weather-dependent emission rate.

The aim of this study is to provide an estimate on the long-term spatial and temporal distribution of airborne pollution originating from the Budapest Chemical Works site. The concentration pattern is investigated as a combination of three factors: wind direction, atmospheric stability and emission rate. Two types of pollutants are differentiated: deflated dust and evaporated gases. Spatial distribution is given for both types, concerning the strongly different emission characteristics.

2. Estimation of dust emission

The major threat from the Budapest Chemical Works site affecting residential areas is the atmospheric transport of dust polluted with DDT. DDT is a pesticide that has been invented in 1948 to prevent insect-related diseases such as malaria and typhus. Despite its remarkable efficiency in disease control, DDT has been shown to have serious environmental effects due to its persistency and bioaccumulation. DDT also affects human health, interfering with the hormone system and causing neuropsychological dysfunction and adverse birth outcomes (Rogan and Chen, 2005; Beard, 2006). Furthermore, as it had been suspected for decades, the relation of DDT to breast cancer was finally proved in 2015 (Soto

and *Sonnenschein*, 2015). Hungary was one of the first countries to ban DDT in 1968. However, it is still detectable in the environment in a very low concentration. A recent survey found DDT and its decomposition products in 2 of 10 urban dust samples in the downtown of Budapest (*Simon*, 2012). A dust analysis survey around the Budapest Chemical Works site was conducted by Greenpeace on October 7, 2015. Although DDT was found only in 1 of 11 samples, its decomposition products, DDD (dichlorodiphenyldichloroethane) and DDE (dichlorodiphenyldichloroethylene) were present in 10 of 11 samples (*Greenpeace*, 2015) (*Fig. 1*). Other chemical tracers proved that the polluted dust originated from the Budapest Chemical Works site, supporting the assumption of a former study that the DDT detected in soil samples originated from dust and not from groundwater transport (*Greenpeace*, 2015; *Wessling*, 2015).

Dust flux from the uncovered ground to the atmosphere can be estimated based on the wind velocity and turbulence characteristics. A widely used approach is to estimate the Q dust emission flux as a cubic function of the U^* friction velocity (*Gillette*, 1978; *Tegen et al.*, 1997; *Takemura et al.*, 2000; *Uno et al.*, 2001; *Xuan*, 2004):

$$Q = a(U^* - U_T^*)U^{*2}, \quad (1)$$

where a is an empirical constant. If the friction velocity is below the U_T^* threshold friction velocity, no dust emission is assumed. The threshold friction velocity is dependent on the particle size and density. By an intercomparison of several parameterizations, the threshold friction velocity was found to be between approximately 0.2 and 0.8 m/s for soil dust particles between 10 and 1000 μm (*Marticorena and Bergametti*, 1995). Smaller particles are more difficult to mobilize with threshold friction velocities reaching 2 m/s. The methods for dust flux estimation were mostly developed for large-scale emissions from deserts and bare soils. In an urban environment, strong microscale turbulence increases dust flux in two ways: (1) it decreases the effective threshold velocity, as turbulent fluctuations can locally generate significantly higher wind speeds than the mean wind; and (2) it increases the dust flux at any given wind speed with a more efficient turbulent upward transport of particles (*Xuan*, 2004). A correction factor proposed by *Xuan* (2004) was applied for the threshold friction velocity to incorporate this effect:

$$U_{T,corr}^* = \frac{U_T^*}{\sqrt{1 + 2\alpha\sqrt{\frac{\sigma_u^2}{U}} + \alpha^2\frac{\sigma_u^2}{U}}}, \quad (2)$$

where U is the mean wind speed, σ_u is the standard deviation of the horizontal wind fluctuation, and α is an empirical constant estimated as $2.5 \text{ s}^{0.5} \text{ m}^{-0.5}$ (Xuan, 2004).

Another important factor affecting dust emission flux is the moisture content of the surface. Most of the polluted dust laid in the area of the industrial site on solid surfaces, therefore, a conservative approach of rapidly drying dust was assumed, and dust emission flux was set to zero only during precipitation events and when snow covered the ground.

3. Estimation of evaporation

In the site of the Budapest Chemical Works, several hundred tons of organic solvents were stored in leaked containers. Vapor of isopropanol, benzene, and PAHs were released into the atmosphere, causing a characteristic odor in the neighboring area and posing a potential threat to human health. PAHs are carcinogenic organic compounds that are present in the atmosphere both in gaseous and aerosol form (Kim *et al.*, 2013). Unlike the rarely occurring DDT, both benzene and PAHs are frequent pollutants of the urban air, originating from fossil fuel burning. Although the lack of emission data and the large urban background pollution makes it impossible to provide concentration estimates, the temporal variation of the evaporation and, therefore, the spatial distribution of long-term pollution can be simulated. Assuming that the ambient air is far from saturation, the Q_E evaporation flux is estimated as the product of the saturation concentration, the U^* friction velocity, and a j mass transfer coefficient (Brighton, 1985). Using the Clausius–Clapeyron relation to obtain the saturation concentration, the following form yields:

$$Q_E = Aj \frac{U^*}{T} \exp\left(\frac{-L_m}{RT}\right), \quad (3)$$

where A is a material-dependent constant, T is the skin temperature of the fluid, L_m is the molar latent heat constant of vaporization, and R is the universal gas constant. In our study, the molar latent heat constant of benzene (34 kJ/mol) was applied and the skin temperature was approximated with the air temperature.

4. Atmospheric dispersion model

Over complex urban environment, the atmospheric dispersion can be best simulated with computational fluid dynamics (CFD) models. However, these models require huge computational cost depending on the applied grid type (Rákai *et al.*, 2014). Therefore, in this study, due to the huge number of model runs, the atmospheric dispersion was simulated in the 6 km vicinity of the site

with a Gaussian atmospheric dispersion model, a widely used approach in local scale long-term air pollution studies (*Cimorelli et al.*, 2005; *Holmes and Morawska*, 2006; *Leelőssy et al.*, 2011, 2014; *Mészáros et al.*, 2012). Concentration was assumed to follow a normal distribution in the crosswind and vertical directions, centered around the downwind axis from the source point. The axis of the plume was gradually elevated from the surface to the median height of the mixing layer. The mixing efficiency, i.e. the standard deviation of the normal distribution was estimated based on the Monin–Obukhov length, L :

$$L = \frac{TU^{*2}}{kgT^*}, \quad (4)$$

where g is the acceleration of gravity, k is the von Karman constant, and T^* is the dynamic temperature, calculated as:

$$T_{day}^* = \frac{-H}{\rho c_p U^*}, \quad (5)$$

$$T_{night}^* = 0.09(1 - 0.5n^2), \quad (6)$$

where H is the sensible heat flux, ρ is the density of air, c_p is the isobaric heat capacity of dry air and n is the fractional cloud cover (*Cimorelli et al.*, 2005). The friction velocity is obtained as a function of the Monin–Obukhov length:

$$U^* = \frac{kU}{\ln \frac{z_{ref}}{z_0} + \Psi_m \left(\frac{z_{ref}}{L} \right) + \Psi_m \left(\frac{z_0}{L} \right)}, \quad (7)$$

where z_{ref} is the wind measurement height, z_0 is the surface roughness (in our study, 10 m and 2 m, respectively), and Ψ_m is the Monin–Obukhov universal function (*Foken*, 2006). Eqs. (4–7) were solved iteratively until convergence. Turbulent wind fluctuations and the consequent standard deviation of the normally distributed concentration field were calculated following the way described in *Cimorelli et al.* (2005).

Meteorological data were obtained from the nearby meteorological station of the Hungarian Meteorological Service at Budapest-Pestszentlőrinc, located 6 km to the southeast from the Budapest Chemical Works site. Hourly synoptic observations and 12–24 hourly radiosonde profiles were used to produce input data for the dispersion model in each hour of the 13-year long period between 2003 and 2015. Mixing layer height was assumed as the lowest altitude where virtual potential temperature exceeded the surface value (*Seidel et al.*, 2010). The temporal period was selected to cover the years from the first report of the pollution (*Dura*, 2003) until the decontamination (*Hevesi*, 2016). Although the emission might have started several years before 2003, the 13-year long period is assumed to be long enough to indicate general dispersion patterns. Missing

data (hours without synoptic observation or no radiosonde profile within 24 hours) occurred in 4.3% of the total period. After excluding missing data, dispersion results for 109,052 hours were evaluated to estimate the long-term load of the neighboring area. In each hour, plumes from 12 equally distributed point sources were superposed to represent the source area. Model results were evaluated on a rectangular grid with 100 m resolution. Without emission data, only the spatial distribution of the pollution can be examined through long-term sensitivity of receptor points. The surface sensitivity maps have been normalized to have a sum of one. Long-term average concentrations are assumed to be proportional to the given sensitivities.

5. Results

Dust emission was estimated at each hour from Eqs. (1)–(2), that show large sensitivity on the value of the U_T^* threshold friction velocity. In the main simulation run, U_T^* was conservatively set to 0.2 m/s. However, results have also been calculated with the more optimistic assumption of 0.8 m/s threshold friction velocity to obtain more focused information on the large wind speed (high dust emission) situations. For comparison, all dust results were normalized with the same value yielding a sum of one in the 13-year mean result of the main case. In the 13-year dataset, 19.6% of the observations provided friction velocities below 0.2 m/s and 15.6% above 0.8 m/s.

Dust sensitivity maps (*Fig. 2*) show a clear domination of dust transport towards the south-eastern direction. Although north-westerly wind is the dominant direction in Budapest (*Fig. 3a*), this result cannot be explained by the simple wind direction statistic that would yield much more isotropic dispersion field with a more southward dominance (*Fig. 3b*). This underlines that dust emission rate is the dominant factor in the dispersion model. While the sensitivities certainly decrease at larger threshold friction velocities, the difference is small, and the general dispersion pattern is very similar. This result can be surprising considering the fact that 64.8% of the observations fell between the 0.2 and 0.8 m/s threshold friction velocities. However, 80% of the total dust emission in the most conservative 0.2 m/s threshold friction velocity setup can be attributed to the observations where friction velocity also exceeded 0.8 m/s. More extremely, 3,750 strong-wind cases of the hourly observations, only 3% of the 13-year period caused 32% of the total dust emission.

The strong southwestward dispersion of dust is due to only a relatively few cases with strong winds that are typically related to cold fronts approaching from the northwest (*Fig. 3a*). The results correspond with the fact that the dust samples to the south and west of the site showed the largest DDD concentrations, however, the samples were not representative (*Greenpeace, 2015*). DDD detection in the dust to the west of the site is not explained by this

model. It could be caused by a short-period strong wind event (i.e., a gust front) from the eastern direction, as easterly winds are relatively frequent in the area (Fig. 3a). Another theory is to contribute this detection to water origin as the sample site lies in the main path of groundwater transport from the factory to the Danube.

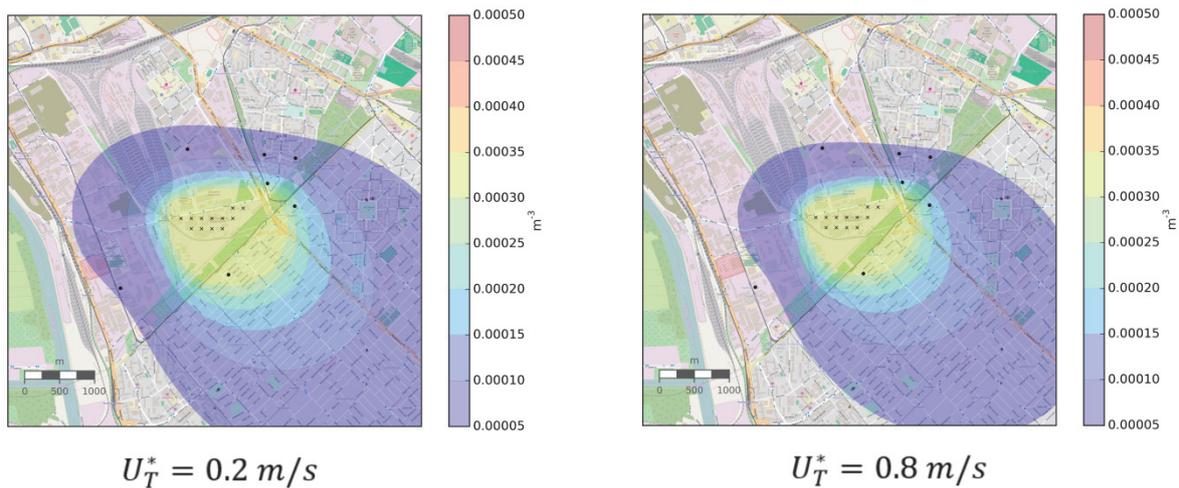


Fig. 2. 13-year mean dust normalized concentrations (sensitivities) at 2 m level, assuming 0.2 and 0.8 m/s threshold friction velocity. Black crosses indicate the source points, and black dots show the DDD detections in soil or dust samples. Background map © [OpenStreetMap](#) contributors, [CC-BY-SA](#)

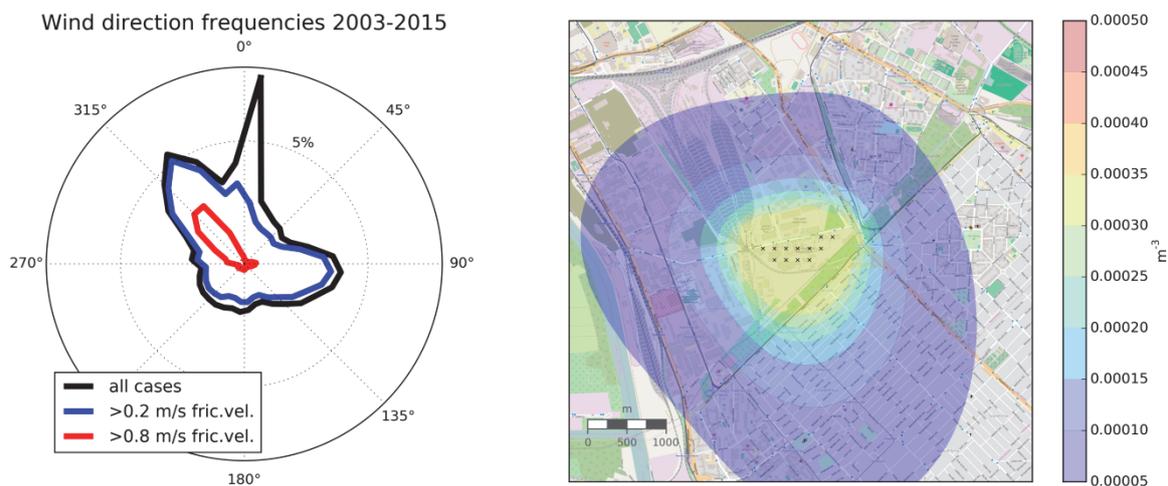


Fig. 3. Left: frequency distribution of wind directions in Budapest-Pestszentlőrinc between 2003–2015 in cases with larger than 0.0, 0.2, and 0.8 m/s threshold friction velocities. Right: reference 13-year mean dust normalized concentrations at 2 m level, assuming constant emission rate. Black crosses indicate the source points. With the consideration of weather-dependent emission rates, this spatial distribution is largely modified (cf. Fig.2. and Fig.4.). Background map © [OpenStreetMap](#) contributors, [CC-BY-SA](#)

The long-term spatial distribution of evaporating pollutants (*Fig. 4*) shows a different image. In this case, the emission rate is only a linear function of the friction velocity and an exponential function of temperature. A combination of these effects yields a south-eastward oriented plume that affects the eastward laying residential area less than the dust transport, but still more than the reference result with constant emission rate would imply. A clear northward direction is also observable related to rare, but extremely warm episodes caused by southerly winds (*Fig. 4*).

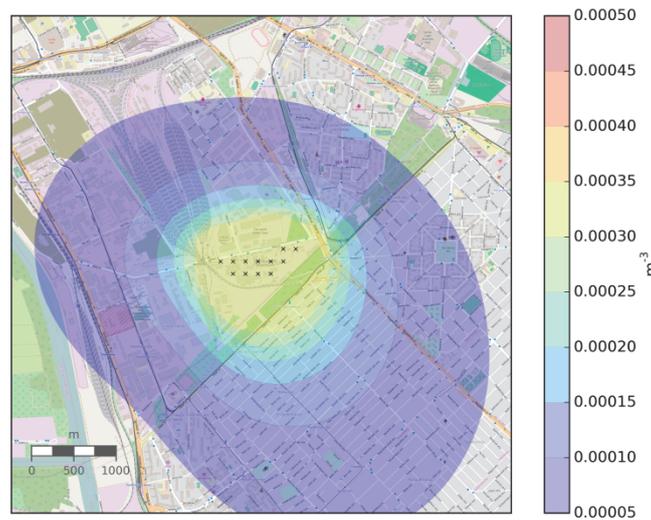


Fig. 4. 13-year mean normalized concentrations (sensitivities) of evaporating pollutants at 2m level. Background map © [OpenStreetMap](#) contributors, [CC-BY-SA](#)

The annual cycle of dust emission flux clearly follows the climatological characteristics of wind velocities, showing a well-defined spring maximum and fall/winter minimum (*Fig. 5*). Annual cycle of evaporation flux has a summer maximum and winter minimum following the temperature cycle. However, evaporation is stronger in the spring than in the autumn due to the generally stronger winds. For similar reasons, the daily cycle of emissions also shows a daytime maximum and a night minimum, according to the temperature and wind speed cycles (*Fig. 5*).

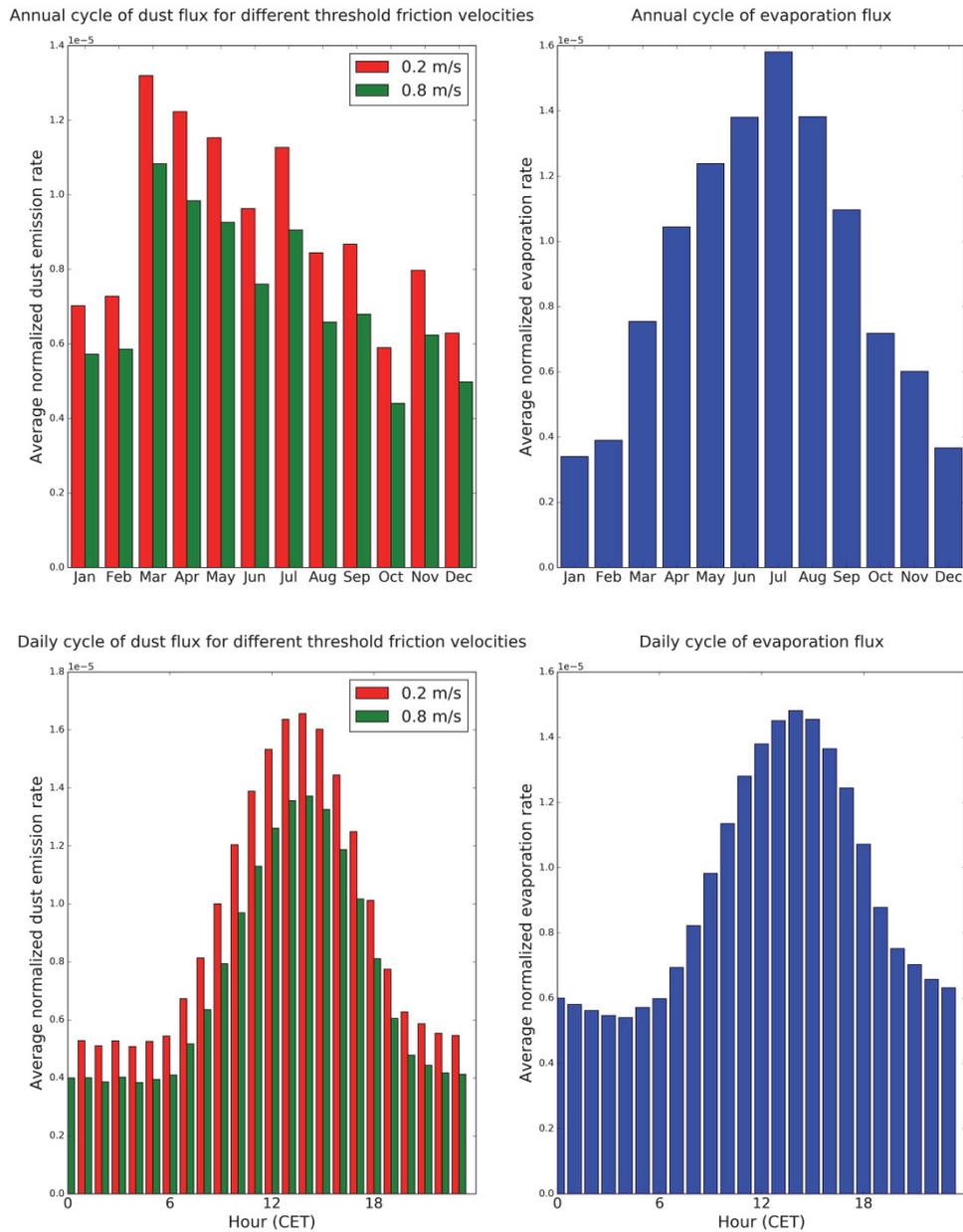


Fig. 5. Annual and daily cycle of normalized dust emission flux at different threshold friction velocities (left) and evaporation flux (right). Strong daytime and spring-summer maximum is observable in both cases.

The interaction of emission and dispersion processes can be interpreted as a favorable relationship to decrease surface air pollution. The emission is small or zero under atmospheric conditions with weak winds and cold nighttime inversions when surface concentrations would become the largest relative to the source term. On the contrary, high emission rate due to warm surface temperatures and strong winds is coupled with efficient turbulent mixing that ensures low surface concentrations relative to the source rate. In the counteraction of emission rate and mixing efficiency, the former factor seems to

be dominant, yielding summertime and daytime pollution maximum (Figs. 6–7). Nights, in general, show very limited emission rate, and thus, negligible pollution compared to daytime cases (Fig. 7). This temporal pattern is the opposite of what is generally experienced in urban air pollution statistics, and points out the importance of correct representation of emission characteristics. The increased risk by the spring-summer and daytime concentration maximum is the high exposure as people spend more time in the open air, and houses are more frequently ventilated. The relatively rare occurrence of significant emission events, however, provides a good chance of prevention in similar cases in the future, based on weather forecasts warning of strong, dry winds, especially if they are accompanied by high temperatures.

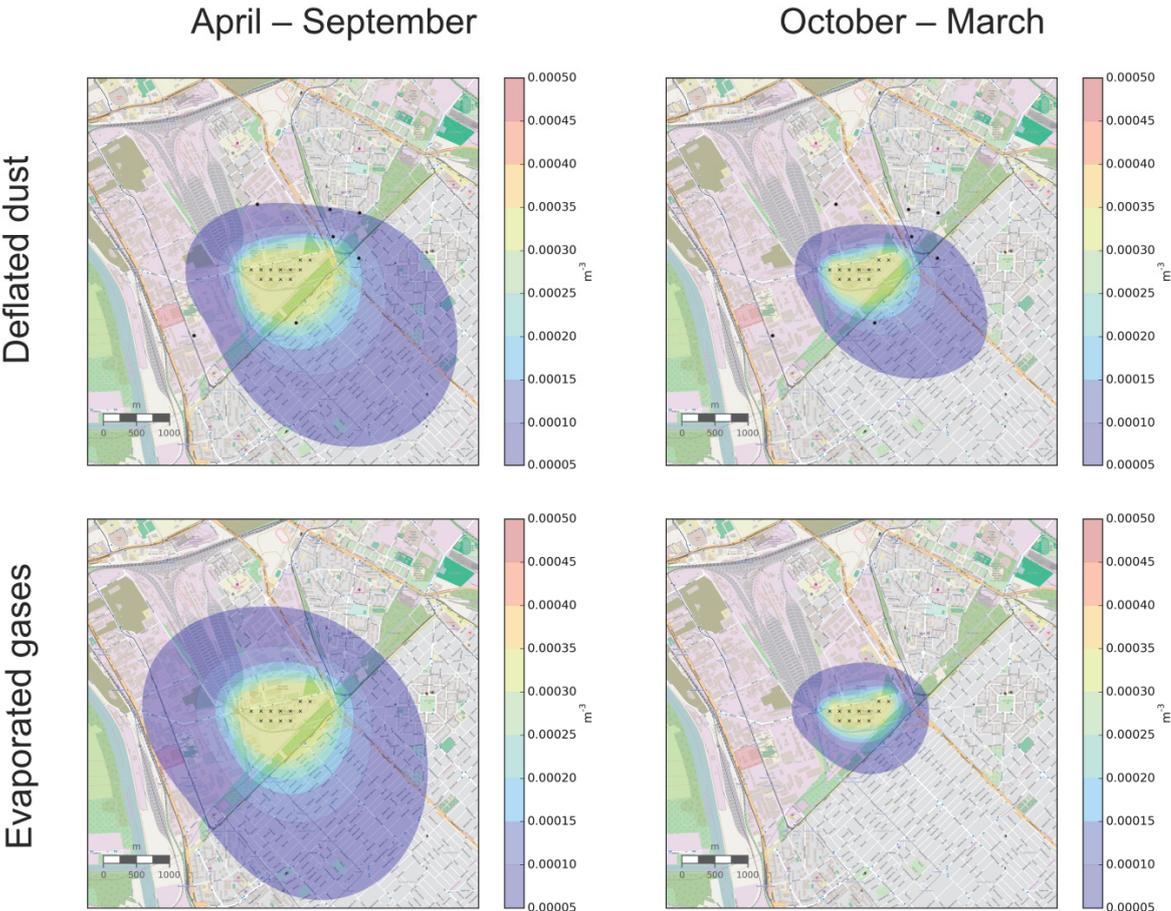


Fig. 6. 13-year mean normalized concentrations of the summer and winter half-years for deflated and evaporated pollutants. The dust pollution of the winter half-year is largely attributable to that of March. Background map © [OpenStreetMap](https://www.openstreetmap.org/) contributors, [CC-BY-SA](https://creativecommons.org/licenses/by-sa/4.0/)

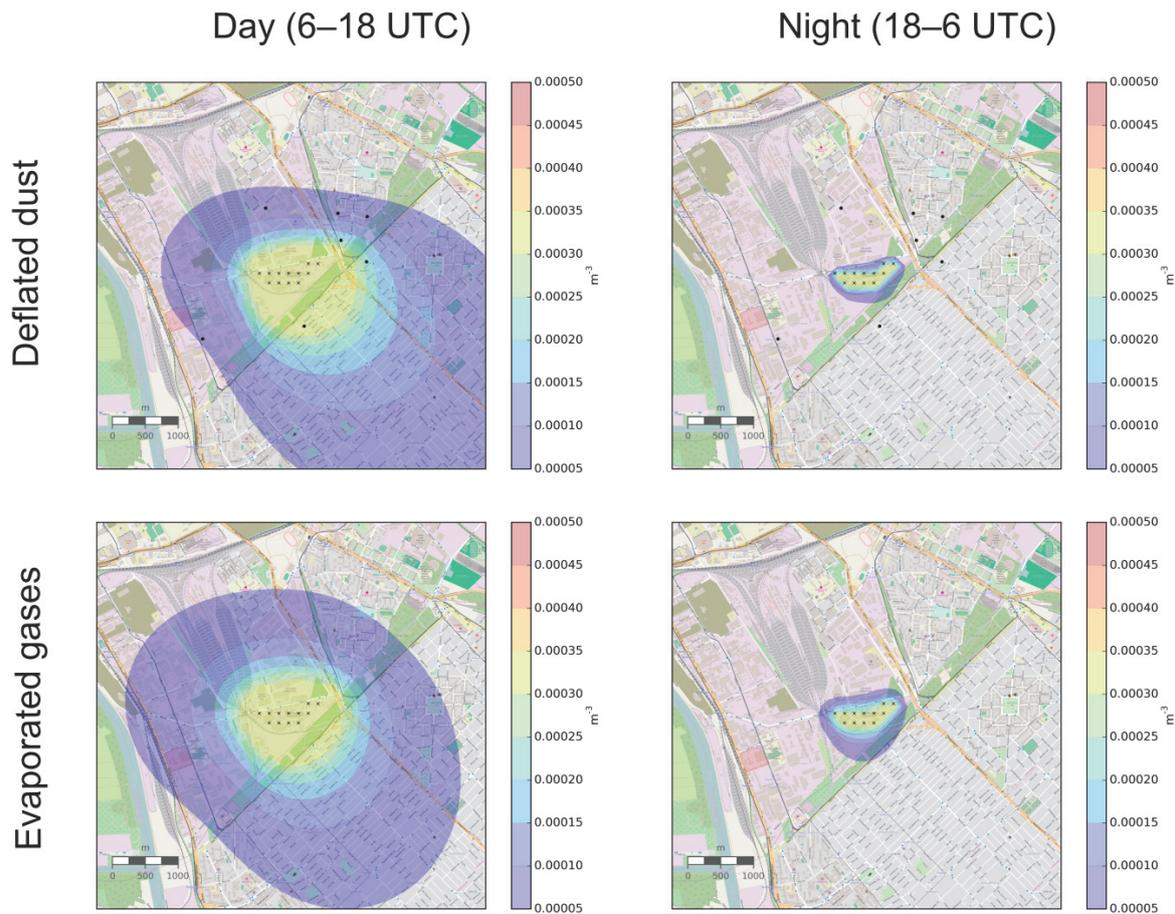


Fig. 7. 13-year mean normalized concentrations of the daytime and nighttime half-days for deflated and evaporated pollutants. Despite the typically lower mixing heights, nighttime concentrations are small due to the decreased emission rate. Background map © [OpenStreetMap](#) contributors, [CC-BY-SA](#)

While the weather-dependent emission rate seems to be the ultimately dominant factor of the pollution, uncertainties and unrepresented effects can alter the results of this model simulation. Most importantly, the moisture content of the dust is an important factor in dust emission estimation. We have conservatively assumed a rapidly drying dust with zero emission only during precipitation, an approximation that seems to be acceptable for the thin dust layer laying on a solid surface. However, emission by strong winds only a few hours after intensive precipitation can still be decreased. This effect would probably decrease the domination of the south-eastern direction as situations with heavy precipitation followed by strong wind are typically related to northerly flows behind cold fronts.

The meteorological observatory is located only 6 km away from the industrial site, in a similar urban-domestic environment and on flat terrain, therefore, its data is assumed to be representative for the release site. However, microscale flow around the buildings can significantly alter dispersion patterns

in the vicinity of the source area, an effect that was partly taken into account by a correction factor. However, as it is generally true for Gaussian dispersion models, results must be carefully interpreted, and their accuracy is limited in a few hundred meters range from the source.

6. Conclusion

The long-term pollution originating from an abandoned industrial site in Budapest, Hungary has been studied with weather-dependent emission rates. In the lack of information on the actual amount of released material, sensitivities have been calculated to examine the spatial distribution and temporal cycle of pollutants. Deflation rate was assumed as a cubic function of the friction velocity, while evaporation rate was estimated as a linear function of friction velocity and an exponential function of temperature. Long-term dispersion of pollutants has been investigated through the 13-year period between 2003 and 2015, using a Gaussian dispersion model based on hourly observations and 12–24 hourly radiosonde data from a meteorological observatory located 6 km to the southeast from the release location.

In case of the dust release, relatively few occurrences of strong winds dominated the long-term dispersion pattern, yielding a distinct south-eastward transport of the plume. The sensitivity of results on a critical parameter of the emission model, the threshold friction velocity, has proved to be small despite the fact that the uncertainty range of the parameter covered almost two thirds of the observations. The spring and daytime maximum of emission rate has been found to be the dominant factor of the dispersion, counteracted by the efficient dilution of intensive turbulent mixing. For evaporated gases, the south-eastward and northward directions were the most affected due to a combination of wind direction distribution and the strongly temperature-dependent emission. Temporal cycle of evaporation closely followed the temperature cycle with summer and daytime maximum.

It has been shown that weather-dependent emissions can significantly alter the results of an air pollution model on local scale. Pollutants from deflation or evaporation origin showed largely different spatial distribution and temporal cycle than what the meteorological conditions and wind direction distribution would have suggested. The results can provide a basis for further measurements and health impact estimates to better assess the pollution risk from the Budapest Chemical Works.

Acknowledgement: The study was largely motivated by personal communications with *Zoltán Barcza*. Authors appreciate the efforts of civil groups and authorities to reveal the most possible information and to decontaminate the site. This work was supported by the National Research, Development and Innovation Office of Hungary (No. K109109 and No. K116506).

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