IDŐJÁRÁS

Quarterly Journal of the Hungarian Meteorological Service
Vol. 117, No. 4, October–December, 2013, pp. 435–450

Comparison of two Lagrangian dispersion models: a case study for the chemical accident in Rouen, January 21-22, 2013

Ádám Leelőssy[1], **Erika Lilla Ludányi**[1], **Márk Kohlmann**[1], **István Lagzi**[2], and **Róbert Mészáros**[1,*]

[1]*Department of Meteorology, Eötvös Loránd University, P.O. Box 32, H-1518 Budapest, Hungary*
[2] *Department of Physics, Budapest University of Technology and Economics, Budafoki út 8, H-1111 Budapest, Hungary*

**Corresponding author E-mail: mrobi@nimbus.elte.hu*

(Manuscript received in final form July 10, 2013)

Abstract—Industrial accidents have been a serious environmental and public health issue for the last decades. Although the development of atmospheric dispersion models was largely motivated by the notorious nuclear catastrophes, simulations are now mostly used in cases of chemical accidents that regularly occur in all parts of the world. In an accidental situation, the accuracy of the results is primarily important for risk management and decision making strategies. However, it largely depends on the meteorological conditions and the quality of input data. A chemical accident happened in a factory in Rouen, France on January 21, 2013. The emitted methyl mercaptan gas caused odor and sickness in densely populated areas, including Paris. The meteorological conditions were rapidly changing in both space and time during the release period, thus the case is particularly challenging for dispersion models and provides a good basis for testing them.

Dispersion of the released methyl mercaptan gas was estimated using the PyTREX trajectory model, developed at the Eötvös Loránd University, and NOAA's HYSPLIT model. The simulation results are in a good agreement with media reports of the polluted areas, and lead to a better understanding of the complex synoptic situation at the time of the accident. Comparison of the results of two models also provided information about the uncertainty of the predictions and pointed out the most important directions for further development of the PyTREX model.

Key-words: atmospheric dispersion, accidental release, HYSPLIT, industrial accident, air pollution, Lagrangian model

1. Introduction

In case of an accidental release of toxic material into the atmosphere, dispersion models provide valuable information for risk management and decision support. In most cases, simulation of the dispersion of pollutants released during an accident is a difficult task because of the complex physical processes occurring in the atmosphere, the importance of fast response, and the lack of information about the details of the release. Computer simulations, based on either Eulerian or Lagrangian (trajectory) approaches are now able to provide fast and accurate estimation about the concentration patterns after an accident.

In the past years, PyTREX, a Lagrangian trajectory model has been developed for regional to continental scale simulation of dispersion of passive pollutants. In this work, we present the PyTREX results for the case of the Lubrizol accident in Rouen, compared against HYSPLIT, a state-of-the-art software, to estimate the uncertainty and show the strengths and weaknesses of our model. The Rouen accident happened under complex meteorological conditions where dispersion models are less reliable and depend largely on the accuracy of their host numerical weather prediction model.

This work aims to provide a case study of the Rouen accident, involving its synoptic meteorological conditions and the consequent dispersion patterns. On January 21, 2013, a gas leak caused a significant release of methyl mercaptan from the Lubrizol factory. Although methyl mercaptan had no health risks, its intense odor could cause nausea and headache. As the dispersion plume crossed densely populated areas, many complaints arrived from the public, and numerous media announcements and reports have been published. Despite the fact that methyl mercaptan gas measurements are not available, these media reports provide information about the affected areas and the intensity of the odor in a particular location, thus the dispersion of the plume can be qualitatively verified.

2. Overview of atmospheric dispersion modeling

Atmospheric dispersion involves multiscale air pollution problems that are treated using different mathematical approaches and modeling tools. Computer simulations have to take into account the horizontal advection of the released pollutant by the mean wind, the horizontal and vertical mixing caused by turbulent diffusion, chemical reactions, wet and dry deposition, sedimentation, and radioactive decay. The wide range of scales and physical processes led to the development of several atmospheric dispersion models that are specialized to the simulation of certain types of air pollution situations.

Microscale models, often referred to as street canyon simulations use a computational fluid dynamics (CFD) approach to solve the governing equations as well as the dispersion equation on a very fine grid around a complex

geometry like a city, a tunnel or an industrial site (*Balczó et al*., 2011; *Di Sabatino et al*., 2008). Sophisticated CFD models like Ansys or OpenFOAM are able to take into account microscale phenomena, the effect of buildings, and turbulence generation on the walls (*Cheng* and *Liu*, 2011; *Yamada*, 2004). This approach provides valuable information about urban air quality (*Vardoulakis et al*., 2003), however, it is not applicable on larger scales due to its large computational cost and the unrepresented physical processes like atmospheric stability and mesoscale wind patterns (*Baklanov*, 2000).

On meso- to macroscale, atmospheric dispersion simulations are based on the output data of numerical weather prediction (NWP) models. Besides the three-dimensional wind field, atmospheric stability characteristics, planetary boundary layer height, and surface parameters are also obtained from NWP results (*Stohl et al*., 2005). Regional and continental scale dispersion models often use the same grid as the host NWP to solve the transport equation. This Eulerian approach has the advantage that meteorological data is obtained without interpolation, complex chemical reactions can be easily taken into account, and the output concentration and deposition fields are directly computed by the model (*Simpson et al*., 2012).

Lagrangian simulations avoid the costly partial differential equation solvers and compute tracer trajectories using the NWP-provided wind field. As the calculation of a few trajectories is very fast, Lagrangian models are able to provide immediate information about the dispersion's direction without calculating concentrations. However, with thousands of trajectories, cluster analyses can be carried out to obtain the concentration field. Turbulent mixing is taken into account with a stochastic random walk method (*Stohl et al*., 2005). Although Lagrangian models require costly interpolation of meteorological data, this approach is particularly suitable for near-source simulations, where numerical diffusion introduces a large error in Eulerian models. This error can be largely reduced by using adaptive gridding that refines the resolution if large gradients are present (*Lagzi et al*., 2009). Coupled modeling systems have also been introduced that use a near-source Lagrangian treatment within a large-scale Eulerian model (*Brandt et al*., 1996).

Lagrangian approach is used in state-of-the-art atmospheric dispersion software like the NAME, HYSPLIT, and FLEXPART models (*Draxler* and *Hess*, 1998; *Stohl et al*., 2005). Besides their worldwide application for environmental studies and risk management, these models provided valuable and accurate information during recent air pollution episodes like the Fukushima accident in 2011 or the eruption of Eyjafjallajökull volcano in 2010 (*Dacre et al*., 2011; *Long et al*., 2012; *Srinivas et al*., 2012; *Stohl et al*., 2011).

The simulation of long-term average air pollution patterns caused by continuous release is a challenge for most atmospheric models. EMEP's Eulerian model provides continental scale forecasts and archive data for most air pollutants' concentration with a special attention on acidic compounds (*Simpson*

et al., 2012). The online coupled dispersion and mesoscale weather prediction model WRF-Chem is a powerful tool for atmospheric dispersion modeling: its Eulerian approach allows the simulation of complex chemical reaction systems, meanwhile, the integrity with an NWP model makes it easy to run detailed simulations in any meteorological situations (*Huh et al*., 2012).

On regional scale, plume models like AERMOD or ADMS are often used to calculate long-term average concentrations caused by a continuous pollutant source (*Holmes* and *Morawska*, 2006; *Silverman et al*., 2007). Plume models assume straight downwind dispersion from the source point and a concentration field with Gaussian distribution in crosswind and vertical direction (*Cimorelli et al*., 2005). Although these models are not reliable in complex weather situations and terrain, their fast runtime makes them optimal for long-term statistical air quality investigations for both normal (*Righi et al*., 2009) and accidental (*Leelőssy et al*., 2011) continuous releases.

In Hungary, an integrated atmospheric dispersion modeling system (AERMOD) and a trajectory and particle dispersion model (FLEXTRA-FLEXPART) are used by the Hungarian Meteorological Service for environmental monitoring and risk management (*Kocsis et al*., 2009; *Steib* and *Labancz*, 2005). The CHIMERE model was also adapted at the Hungarian Meteorological Service for operative mesoscale air quality forecast in Budapest (*Baranka* and *Labancz*, 2009). At the Paks Nuclear Power Plant, the RODOS decision support system provides a Lagrangian trajectory model for regional to continental scale simulations. The SINAC program system was developed to follow the consequences of radioactive releases of a hypothetical nuclear accident (*Földi et al*., 2010). A multiscale Lagrangian and Eulerian dispersion model, TREX has also been developed at the Eötvös Loránd University for the area within 30–500 km from the power plant (*Mészáros et al*., 2010). For larger scales, the extended PyTREX trajectory model has been developed. Local scale CFD simulations are carried out at Budapest Technical University and Eötvös Loránd University using Fluent, Miskam, and OpenFOAM models (*Balczó et al*., 2011; *Goricsán et al*., 2004).

3. Model description

3.1. The HYSPLIT model

In the present work, we used HYSPLIT and PyTREX models to simulate the consequences of the industrial accident in Rouen. HYSPLIT is a widely used Lagrangian dispersion model developed by the National Oceanic and Atmospheric Administration Air Resources Laboratory (NOAA ARL). Its worldwide applications cover various forward and backward simulations from meso- to continental scale (*Challa et al*., 2008; *Koracin et al*., 2011; *Long et al*., 2012; *McGowan* and *Clark*, 2008; *Shan et al*., 2009). HYSPLIT calculates

single trajectories based on meteorological fields provided by the Global Data Assimilation System (GDAS) database. Particle motion in each timestep is defined as a sum of an advective and a turbulent component (*Draxler* and *Hess*, 1998). The advective motion is obtained directly from the wind field, however, vertical turbulent wind fluctuations are computed using Hanna's parameterization based on stability characteristics defined by the Monin–Obukhov length (*Draxler* and *Hess*, 1998; *Moreira et al*., 2011). While large scale turbulence is estimated with a random walk method, small scale turbulent diffusion is calculated with a puff approach: each particle has a horizontal extent with a Gaussian concentration distribution, which broadens according to the local turbulence intensity. Concentration field is given as the superposition of concentration fields of all particles.

3.2. The PyTREX model

PyTREX is a continental scale trajectory model developed at the Eötvös Loránd University. It computes single particle trajectories based on meteorological data provided by short-range forecasts of the Global Forecast System (GFS). GFS is initialized in every 6 hours and provides output fields for every 3 hours, thus the first and second timestep of each model run was used to create a continuous 3-hourly forecast database for archive situations. Forecast outputs were preferred against analyses in order to gain advantage of GFS parameterizations that provide derived quantities such as turbulent surface fluxes or precipitation patterns. GFS grid has 0.5-degree spatial resolution from which data is obtained for any point with linear interpolation in both space and time. For compatibility with the GFS outputs, PyTREX uses spherical coordinate system in horizontal and pressure system in vertical direction. Meteorological and user-defined input data of PyTREX are presented in *Table 1*.

Table 1. Input data requirements of the PyTREX trajectory model

Release data and simulation setup	**Meteorological data (GFS)**
Release location(s)	Geopotential on main pressure levels
Release height(s)	Wind components on main pressure and near-surface levels
Release time(s) and length(s)	Temperature on main pressure and near-surface levels
Simulation duration	Surface pressure, temperature
Total released mass from each location	Surface height above ground level
Number of trajectories from each location	Planetary boundary layer height
Minimum computational timestep	Surface momentum and heat flux
Halftime of radioactive decay	Mixing ratio on main pressure and near-surface levels

PyTREX trajectories are calculated using a linear scheme from the superposition of advective and turbulent motions:

$$\frac{d\underline{r}}{dt} = \underline{v} + \underline{v}_t, \tag{1}$$

where $\underline{v}$ is the vectorial sum of the horizontal wind and the vertical motion, $\underline{v}_t$ is the vector of turbulent fluctuations, and $\underline{r}$ is the position of the particle. While $\underline{v}$ is directly obtained from GFS outputs, $\underline{v}_t$ is calculated using the Langevin equation (*Stohl et al*., 2005):

$$dv_{t,i} = -v_{t,i}\frac{dt}{T_{Li}} + \sigma_i\sqrt{\frac{2dt}{T_{Li}}}\zeta(0,1), \tag{2}$$

where $v_{t,i}$ is the *i*th component of the turbulent velocity vector, T_{Li} is the Lagrangian timescale representative for the *i*th direction, σ_i is the turbulent fluctuation of the *i*th component of the wind vector, and $\zeta(0,1)$ is a random number from a standard normal distribution, generated with the Mersenne Twister algorithm of Python's *random* module.

The TLi Lagrangian timescales and σ_i turbulent wind fluctuations are estimated using the Monin–Obukhov theory, thus we need to compute the atmospheric stability parameter z/L (*Draxler* and *Hess*, 1998):

$$\frac{z}{L} = \frac{z_1 \cdot k \cdot g \cdot T*}{T_1 \cdot u*^2}, \tag{3}$$

where z is the height above ground, L is the Monin–Obukhov length, k is the von-Kármán constant, and g is the gravitational acceleration. Besides constants and surface parameters, PyTREX uses the temperature data T_1 of z_1 height, the first level above ground in the meteorological dataset (80 m for GFS data).

Friction temperature $T*$ and friction velocity $u*$ are calculated from surface heat and momentum fluxes:

$$u* = \left(\frac{\sqrt{\overline{\rho u' w'}^2 + \overline{\rho v' w'}^2}}{\rho}\right)^{0,5}, \tag{4}$$

$$T* = -\frac{H}{\rho_f c_p u*}, \tag{5}$$

where $\overline{\rho u' w'}$ and $\overline{\rho v' w'}$ are surface momentum fluxes and H is the surface heat flux. Both momentum and heat flux data is directly obtained from GFS outputs. The air density ρ and air density on surface ρ_f are calculated from the temperature field using dry air assumption. Accordingly, c_p is the specific heat of dry air.

Based on the stability characteristics presented in Eqs. (3–5) and the planetary boundary layer height provided by GFS, the velocity fluctuations and Lagrangian timescales are obtained through Taylor's parametrization, which was set up in a way presented by *Moreira et al*. (2011). The computational timestep *dt* is defined as the tenth of the minimum of Lagrangian timescales (*Stohl et al*., 2005). However, in order to reduce the computational cost for near-surface trajectories, a minimum timestep can be defined that also gives a lower boundary for Lagrangian timescales.

Besides drawing single trajectories, PyTREX calculates concentration field on a three-dimensional rectangular grid based on the density of trajectories crossing the specified grid cell during a certain time period.

4. Synoptic situation during the Rouen incident

On January 21, 2013, a chemical accident happened in a factory of the Lubrizol company located in Rouen, northwestern France. The firm announced that a significant amount of non-toxic methyl mercaptan gas had been released from approximately 07 UTC (http://www.paris-normandie.fr/article/actualites/en-direct-fuite-de-mercaptan-chez-lubrizol). Although no health risk was identified, an unpleasant smell spread across northwestern France after the accident, reaching Paris at the following night. Media announcements reported serious complaints of odor from several districts of the capital. Odor caused by methyl mercaptan gas was also reported from Normandy and Southeastern England (*Fig. 1*) (http://www.bbc.co.uk/news/world-europe-21147361).

Looking at the map of northwestern France (*Fig. 1*), it might be confusing that odor was reported within 24 hours from largely different directions from Rouen, including Paris, which is located to the southeast from the location of the accident, and also from England, to the northwest of the factory. Furthermore, despite that the accident happened only 120 km away from the capital, it took more than 12 hours for the plume to reach Paris. These unusual dispersion patterns were caused by a complex synoptic situation involving a significant shift in the wind direction within a short time period.

On January 20, 2013, two dominant processes were detectable that would determine the spreading of the emitted material. The first synoptic object was a mature trough above the Mediterranean – North African region, with a corresponding low pressure system above the western basin of the Mediterranean Sea (*Fig. 2* (a)-(d)). This low pressure system was severed off

into a bi-central system by the inertia of the cold air arriving at the rear of the trough, along the western coastlines of France (*Fig. 2* (a)). The primary low remained above the Mediterranean, and was being advected eastward with the rest of the trough, while the secondary low (object A), gaining enhanced circulation by baroclinity was advected towards Northern France.

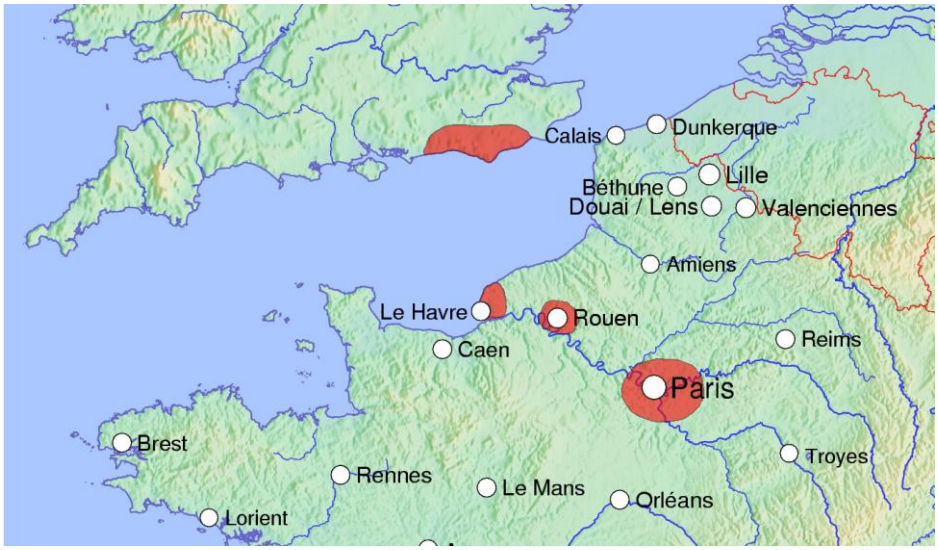

Fig. 1. The most affected areas based on media coverage after the Lubrizol chemical accident in Rouen, January 21-22, 2013.

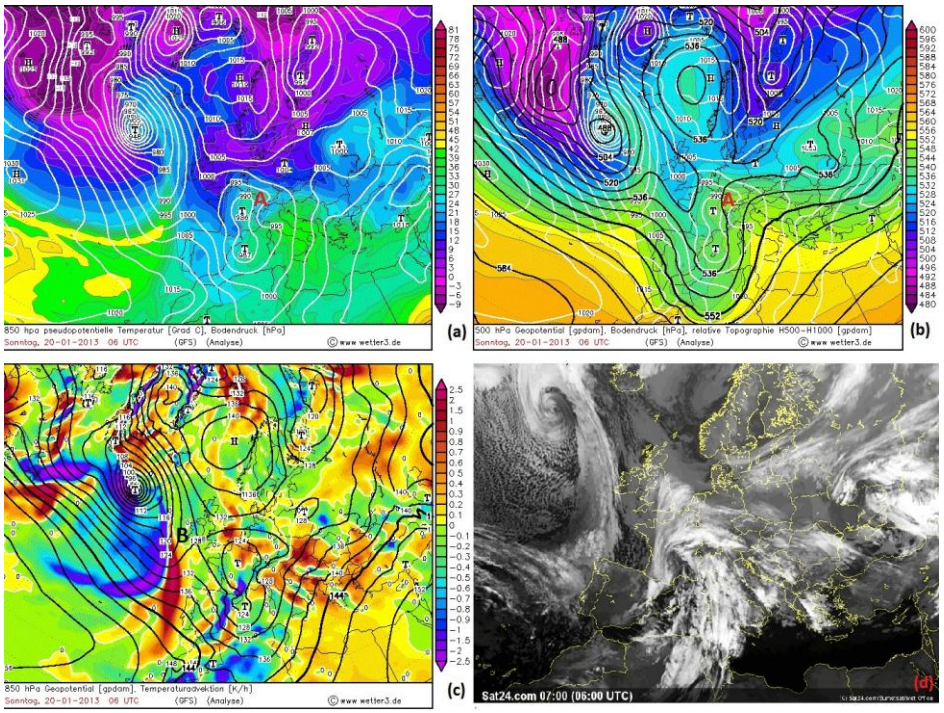

Fig. 2. GFS output (a)-(c) and infrared satellite image (d) at 06 UTC, January 20, 2013. (a) Equivalent potential temperature and MSLP (EPT850) (b) 500 hPa height (gpdam), MSLP and 500/1000 ReTop, (c) Height (gpdam) and TA at 850 hPa. Courtesy of wetter3.de and sat24.com, respectively.

The second main synoptic process was a rapidly deepening trough above the Atlantic, characterized by strong winds and cold advection on lower levels (*Fig. 2* (c)). The leading edge of this trough reached the Rouen region with a cutoff low on the ground level (object B) (*Fig. 3*). Between 18 UTC, January 20, and 12 UTC, January 21, the two low pressure systems started merging in a circular motion (Fig. 3) with the first system (object A) following the streamlines of the second low (object B). At 06 UTC, January 21, the two main lows could be located at Bretagne and Southern England creating the rotating flow that would spread the emitted material southeast and northwest of the facility at the lower levels of the troposphere (*Fig. 4*, *Fig. 5*).

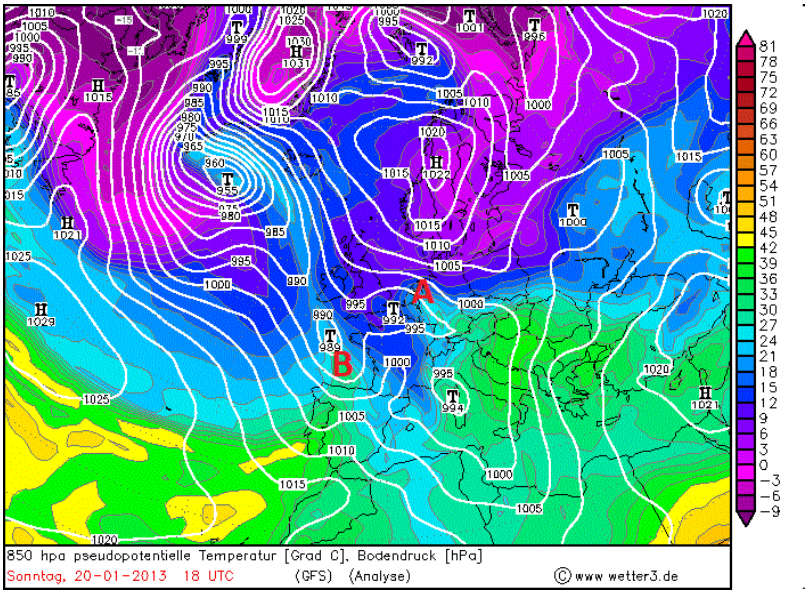

Fig. 3. GFS output on 18 UTC, January 20, 2013, EPT 850. The two stream defining lows begin to merge above the Channel. Courtesy of wetter3.de.

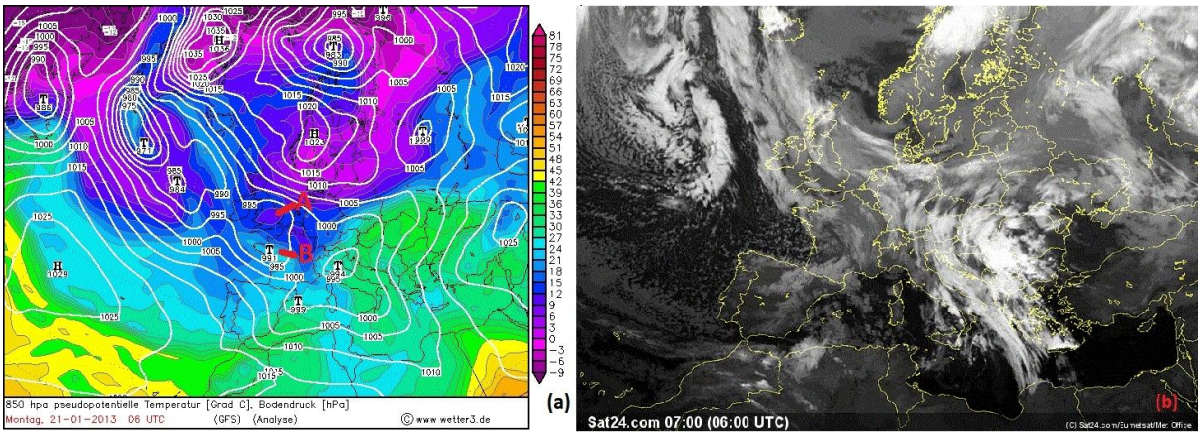

Fig. 4. GFS output (a) EPT850 and infrared satellite image (b) at 06 UTC, January 21, 2013 depicting the synoptic setup shortly before the accident. The two merging lows are clearly visible on (a), above Bretagne and Southern England. On (b), only the rotating field of the southern system is visible. Courtesy of wetter3.de and sat24.com, respectively.

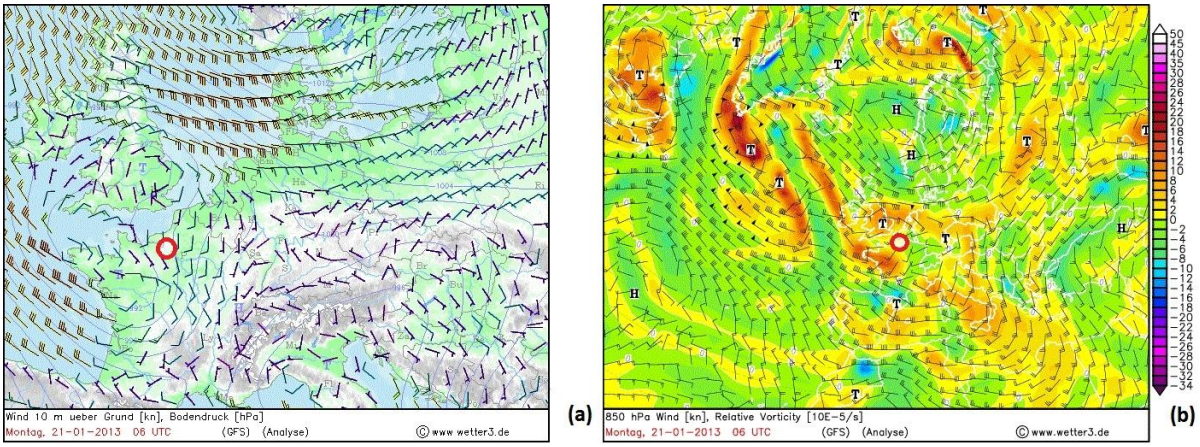

Fig. 5. GFS output at 06 UTC, January 21, 2013. Wind at 10 m (a), wind and vorticity at 850 hPa (b), depicting the bi-central rotating flow at the marked location of the accident. Courtesy of wetter3.de.

Synop reports also show a gradual shifting of the mean wind from northwesterly (18 UTC, January 21) to southeasterly direction indicating the presence of the rotating flow on the ground level. The temperature field did not significantly change due to the overcast nocturnal sky.

5. Dispersion model results

Two trajectory models, HYSPLIT and PyTREX were used to simulate the dispersion of the plume released from the Lubrizol factory during the incident. Besides understanding the pollution patterns reported in the media, our investigation aimed to compare the model results in this complex synoptic situation in order to estimate the uncertainty of trajectories and the concentration field.

The same release data was used for both model runs. Assuming a 24-hour long continuous release from 50 m height, 20 400 trajectories were calculated with evenly distributed starting time during the release period. The number of trajectories was given by default in HYSPLIT, and the same value was used in PyTREX for comparable results. As the exact quantity of the released material was not known, unity total released mass was assumed for the simulation. No wet and dry deposition was taken into account, which is a good assumption for mercaptans. In PyTREX, output concentration map was produced with a 0.25 degrees horizontal and 100 m vertical resolution. Both model calculated one-hour average concentrations for each location.

HYSPLIT results clearly show the wind shift during the release period: in the first 7 hours of the accident, the plume is advected by weak southern wind over the La Manche channel (*Fig. 6*). Between the 7th and 15th hour of the incident, the wind became stronger and changed to northwesterly direction,

which forced the plume back to Northwestern France, reaching again the source region and also Paris. The weak dispersion towards England during the first hours explains the delay between the accident and the pollution reports in Paris, as well as the fairly high intensity of the odor that reached the capital. After the 15th hour of the accident, the wind turned to southeasterly again, and the plume spread towards Southern England, reaching the country approximately 24 hours after the beginning of the release.

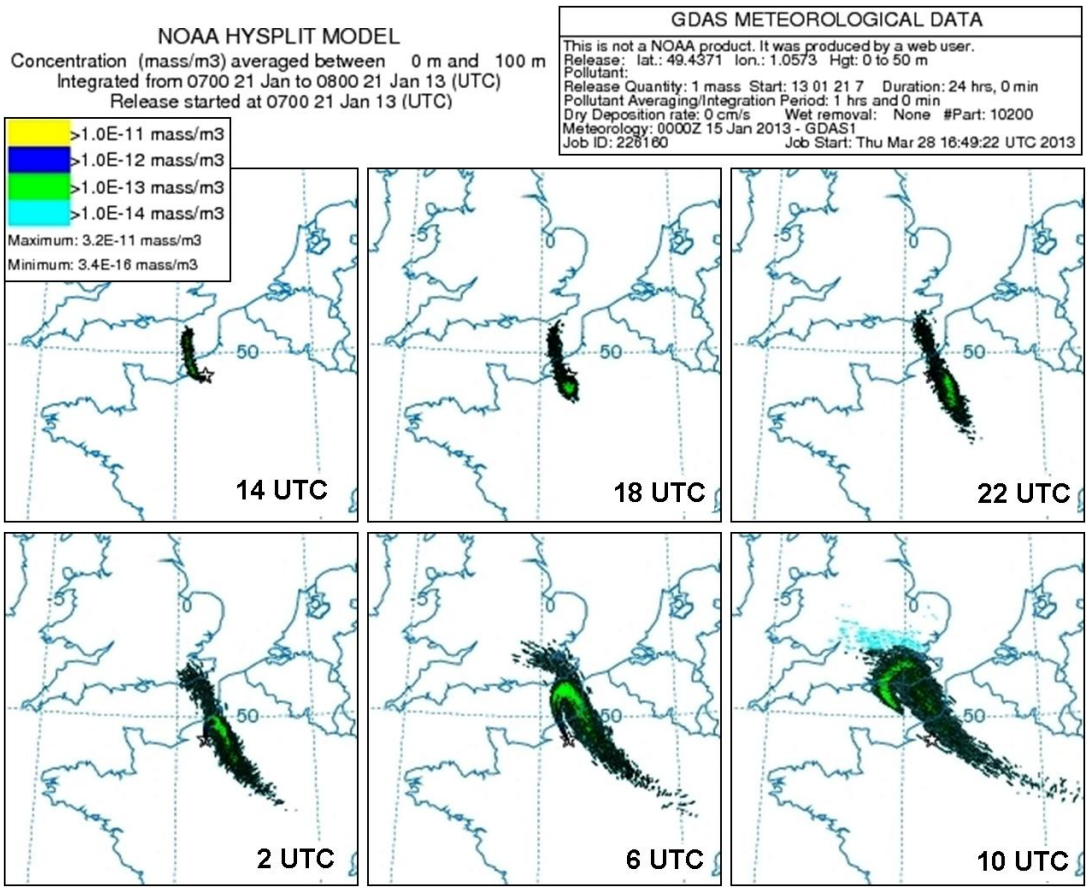

Fig. 6. HYSPLIT surface concentration field between 14 UTC, January 21 and 10 UTC, January 22, 2013. A 24 hours long continuous release was started at 07 UTC, January21. 360° change of wind direction is observable that allowed the plume to reach Central France.

The PyTREX results also well demonstrate the rapid wind shift (*Fig. 7*). The affected areas by the plume are in good qualitative agreement with HYSPLIT's results despite the different meteorological data and physical parameterizations of the models. We note that 27 hours after the beginning of the accident, both models expected that the plume would reach London. In fact, there are a few reports about odor complaints in London, thus the diluted pollutant could reach the city in a fairly high concentration (http://www.dailymail.co.uk/news/article-2266383/Smelly-gas-cloud-factory-Rouen-travels-Channel-France-Kent.html).

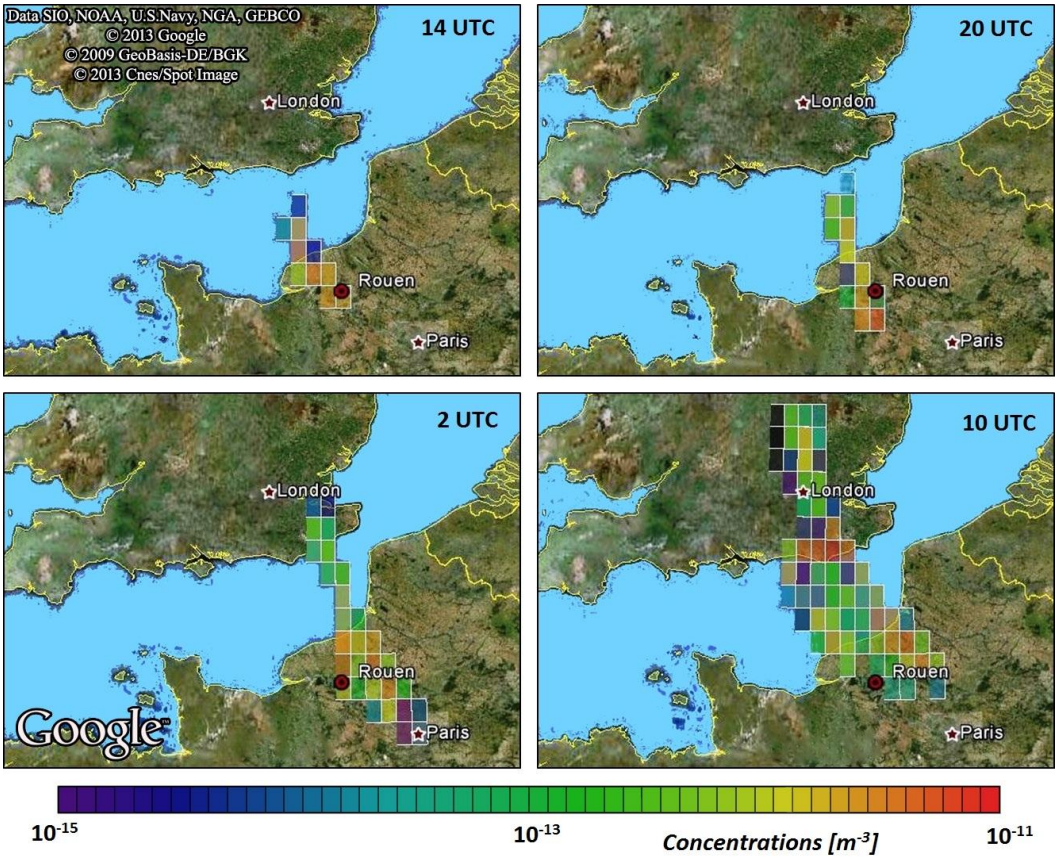

Fig. 7. PyTREX surface concentration field between 14 UTC, January 21 and 10 UTC, January 22, 2013. A 24-hour long continuous release was started at 7 UTC, January 21. Results show a good agreement with HYSPLIT's output.

In *Fig. 8*, three trajectories are presented, started in the 1st, 6th, and 16th hours of the accident. It can be seen that the pollutants spread towards Paris only within a few-hour long time period, before and after which the wind forced the plume to northern, northwestern direction.

Fig. 8. PyTREX trajectories started from Rouen at 07 (red), 13 (yellow), and 22:30 (green) UTC, January 21, 2013. Meteorological conditions allowed the plume to spread towards Paris only within a few-hour long time period.

In order to compare the concentration estimates provided by the two models, the maximum one-hour average concentration was obtained for six locations (*Table 2*). It can be seen that the values are largely different, but remain within the same magnitude for most of the locations, however, close to the source, one magnitude difference is present.

Table 2. Maximum concentration in selected locations based on two models' simulations

Location	Coordinates		Max. concentration [10^{-13}/m^3]	
	Latitude [°]	Longitude [°]	HYSPLIT	PyTREX
Rouen	49.375	1.125	149.50	34.30
Gaillon	49.125	1.375	3.31	24.70
Dieppe	49.875	1.125	8.02	8.88
Paris	48.875	2.375	1.50	6.76
London	51.375	–0.125	1.39	2.93
Bristol	51.376	–2.625	0.82	1.39

It can be concluded that the models are in a good agreement in determining the direction of the dispersion and the affected areas by the plume. The high uncertainty in concentration values might occur from the largely different turbulence treatment of the models: while HYSPLIT uses a mixture of random walk and Gaussian turbulence models, PyTREX performs a 3D random walk turbulence simulation. Based on this knowledge, PyTREX probably underestimates the near-source concentration, because it averages the density of trajectories for a 0.25×0.25 degree cell. Although no measurements are available for methyl mercaptan gas, public complaints of odor can be used to verify the models (http://www.lemonde.fr/planete/article/2013/01/22/fuite-de-gaz-a-lubrizol-mobilisation-maximale-mais-prevention-floue_1820793_3244.html). While Paris was largely affected by the plume, only a few complaints are known from London. HYSPLIT expected a similar concentration value in both cities, which is unlikely.

The uncertainty of the results might also be caused by different meteorological data: while HYSPLIT uses analyses fields, PyTREX is based on short-range forecast files with derived surface parameters.

6. Conclusion

The chemical accident in Rouen on January 21–22, 2013 happened in a complex synoptic situation with rapidly changing wind direction. Two trajectory models were used for the simulation of the dispersion in order to

understand the effect of an interplay of complex meteorological conditions, as well as to compare the model results. The results of PyTREX, a three-dimensional trajectory model developed at the Eötvös Loránd University were compared against the output of HYSPLIT, a widely used atmospheric dispersion model developed by NOAA.

The pollution affected areas in largely different directions because of the rapidly changing wind governed by a multi-centered low pressure system located above Northern France and Southern England. During the release period, a 360° turn of the wind direction was observable, as the dominant southerly wind turned into northwestern direction for a few hours, which allowed the plume to return above Northwestern France and reach Paris. Later, as the wind turned back to southerly direction, the plume crossed the Channel and affected Southern England and London.

Despite the complex synoptic situation, the different meteorological input data, and the fast changing conditions, HYSPLIT and PyTREX results were in a good agreement regarding the dispersion and the polluted areas. Concentration values in selected locations showed large differences, but remained within the same order of magnitude in most cases. PyTREX largely underestimated the near-source concentrations, while HYSPLIT provided unlikely similar results for Paris and London. Uncertainty between models is probably caused by their different turbulence treatment, which requires more sophisticated investigation and verification against measurement data.

The case study of the Rouen incident showed that PyTREX provides reliable results of dispersion patterns even in a complex synoptic situation, however, concentration values have one order of magnitude of uncertainty between the two tested software. Parallel usage of the two models, as well as adjusting parameterizations based on measurement data can largely improve atmospheric dispersion simulations to provide valuable information for risk management in a case like the Lubrizol incident in Rouen.

***Acknowledgements*–**Authors acknowledge the financial support of the Hungarian Research Found (OTKA K81933, K81975, K104666, K109109, and K109361), the *Zoltán Magyary* Postdoctoral Fellowship, the European Union, and the European Social Fund (TÁMOP 4.2.4.A-1).

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