Measurement based source term reconstruction of a radioactive release With use of a dual dispersion model



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Abstract

Everyday, worldwide measurement networks for radioactivity sample the air to detect radioactivity. Sometimes stations sample radioactive materials, while the source is unknown. Because of such incidents we propose and analyze a so called source term reconstruction method, in order to find the most likely source location, release time and duration and released quantity. For this the Dutch National Institute for Public Health and the Environment (RIVM) provided the opportunity to work with an air dispersion model, which uses current weather and weather predictions originating from the Dutch national weather service (KNMI). We propose a source term reconstruction method for an instantaneous release, which is based on a combination of previous literature together with an own contribution. The performance of the method is tested for different variables as sampling locations, weather conditions and measurement and model errors. Finally a real case is used to show that despite of measurement and model errors, the proposed reconstruction method is still able to find possible source terms closely to the actual source term.

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Nomenclature

Roman Symbols

- D() An arbitrary distance operator.
- *f* Chapter 3: Forcing term of the dispersion model.
- *f* Chapter 4: Root Mean Squared Error reduction factor which defines the solution.
- *G* Matrix existing of all dual concentrations for each measurement and each possible combination of location, time and nuclide.
- H() Forward operator representing the air dispersion model.
- *K* Diagonal matrix with diffusion coefficients in three directions.
- *L* Linear operator representing the dispersion model.
- L^* Linear operator representing the dual dispersion model.
- *M* The number of measurements.
- *p* Forcing term of the dual dispersion model.
- *Q* Quantity of release.
- \hat{Q} Computed quantity of release by the method.
- \vec{r} Three dimensional vector in space.
- t Time
- \vec{u} Three dimensional wind vector.

Greek Symbols

 ε^{mod} Model error

ϵ^{meas}	Measurement error
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- λ Deposition and decay parameter.
- μ Measured concentration of radioactive materials in the air.
- ∇ Gradient vector.
- ϕ Concentration field of the simulated release from the actual source term.
- ϕ_m^* Concentration field of a similated retro plume from measurement station *m*.
- σ Source term: concentration field at moment of release.
- *v* Measurements without errors

Subscripts

- 0 Real source term.
- *m* Measurement.
- *p* For each possible source location *p*, t_p is the best moment according to the method and shows a quantity of release given by \hat{Q}_p .

Other Symbols

 \mathcal{D} Domain of an operator.

Acronyms / Abbreviations

- $Corr_0(\vec{a}, \vec{b})$ Correlation forced trough zero of vectors \vec{a} and \vec{b} .
- *PSL* The set of Possible Source Locations.
- *psl* Location in the Possible Source Location grid.
- *Red*() Root Mean Squared Error reduction.
- *RMSE*() Root Mean Squared Error.

Chapter 1

Introduction

Nowadays nuclear power plants are still crucial in the production of electricity. About 450 nuclear power plants positioned around the world generate around 11% of the world's electricity [1]. Although working with nuclear materials involves risks, because of the many safety precautions, nuclear power plants are safer than the current mayor energy sources as coal and oil[2]. Most of the few nuclear accidents in the past were caused by (a combination of) technical or human failures, resulting in damaged nuclear fuel materials due to an inadequate cooling system [3].

One known example is the accident at the Fukushima Dai-ichi nuclear power plant on the northeastern coast of Japan, after the earthquake and tsunami on March 2011. Due to the earthquake, the external power of the reactors was cut off. To make it worse, a tsunami disabled the backup generators and crippled the reactor cooling system [4]. Overheating fuel in an operating reactor led to hydrogen explosions and damaged three other reactor buildings. Fuel in three of the reactor cores melted, and radiation was leaking from the damaged reactors. Those reactors contaminated a wide area surrounding the plant and forced to evacuate nearly half a million residents. Fortunately, no deaths or serious injuries was caused by radioactivity, however many were killed by the tsunami [5]. There was a lot of chaos and fear during these unfortunate events and therefore it is important to predict the effects of nuclear releases to calm the panic and to take adequate measures.

For similar events, scientists have developed dispersion models to forecast the effect of a nuclear accident. Depending on the expected concentrations of radioactivity over space and time, decisions about measures can be formulated, for example an evacuation. Even if such a dispersion model is available, the greatest problem is to collect all information about the source, the exact location, period of release, released materials, quantity of release, release height and other relevant information. This is also called the source term. However, in many cases, such as the nuclear accidents in Chernobyl (1986) and Fukushima (2011), the source term is not exactly known. Often the exact quantity and released materials are not known. Sometimes, as with Fukushima, even the exact location is uncertain, even though it is located within a restricted area. The lack of such important information may result in large uncertainty in the predicted pollutant dispersion. So the source term is crucial for a good prediction of potential concentration of radioactivity in the air and on the ground.

An example of interesting measurements of radioactive materials in the air without a known source term, was the ruthenium mystery in Europe during October 2017. For a couple of weeks, multiple stations around Europe measured small increments of radioactive materials in the air, as is shown in Figure 1.1, while the origin remained unknown[6]. Those measurements raise questions across Europe about the source location and what exactly happened. Multiple organizations such as IRSN in France and the federal office for radiation protection in Germany, BfS, carried out simulations to determine the most likely source term which could have lead to the measured quantities. Those results suggested that the release probably originated between Volga and Ural regions in Russia. However, Russian scientists and engineers pointed out that it is unlikely that a release of ruthenium in the Urals would be detected after such an distance. This mystery has become a political issue and the answer has never been found.

Due to the ruthenium mystery and earlier events as Fukushima, the Dutch National Institute for Public Health and Environment (RIVM) would like to have the ability to compute a source term based on measurements, since no direct method is available yet. Even though air measurements, weather data and dispersion models are available. Therefore, in this thesis a method is proposed to determine the likelihood of different unknown source variables based on air measurements, in order to find the most likely source term of the unknown nuclear release. In this thesis, the reconstruction of the source location, time and quantity of release is the main focus. Furthermore, the proposed method will be analyzed for different variables. One important variable is the positioning of the sampling stations. Although the locations can not be changed, it is interesting to know how the method depends on those locations, since the position of the sampling stations highly influence the measured quantities which are input for the method and thereby affects solutions of the reconstruction method. Another interesting variable is the weather. Even though many different sensors are able to detect the current weather, not all weather properties can be detected and thereby the actual weather can not be simulated perfectly. Moreover, the actual weather conditions are in general only recorded after a couple of hours, which will have consequences for the expected



Fig. 1.1 Measurements of Ruthenium in Europe in September and October of 2017. Values are given in mBq/m³ [7].

dispersion of pollutants in the air. Besides the uncertainty in weather conditions, as for every mathematical model, dispersion models generate errors during the computational process. The same applies for measurements from sampling stations. Because of the existence of model and measurement errors, the effect of those errors on the reconstruction method are analyzed in this thesis.

To start, there is a small recap about radioactivity in the next chapter. In chapter three the mathematical problem is stated and some previous work in source term reconstruction are discussed. After that, the dispersion model available at the RIVM is discussed in detail, the reconstruction method is presented and mathematically analyzed for measurement errors and change of computed release quantity over time. The goal of the fifth chapter is to determine the sensitivity of the method to sampling locations and number of sampling stations, for

incomplete weather information and for measurement and model errors. The sensitivity to positioning of sampling stations and weather conditions are tested by synthetic cases, for which the simple wind fields are generated manually and measurements are modelled by a dispersion model. Furthermore, the sensitivity to measurement and model errors is tested with the real European Tracer EXperiment called ETEX1. The advantage of the chosen cases is that the source terms are known exactly and with that, the performance of the method can be compared with the actual source terms. To complete this thesis, the last chapter will contain conclusions and recommendations for future work.

Chapter 2

Radioactivity

2.1 What is radioactivity?

All materials are made up of many atoms, consisting of a unique combination of protons, neutrons and electrons. However, there are combinations of protons and neutrons which are unstable. An unstable atom nucleus will decay over time [3]. During this decay process energy is released for instance in the form of the alpha radiation (release of an heavy alpha particle), beta radiation (release of a beta particle) or gamma radiation (release of a high energy photon).

The danger of radiation is that it could damage the DNA in the cell, which as a result could kill the cell. Although cells have mechanisms to repair DNA, there are cases the cell can not be repaired or is repaired wrongly, which could result in mutations. This could lead to development of health problems, such as cancer. Since photons does not have mass in contrast to alpha and beta particles, alpha and beta particles have more energy and are more destructive along their way. However alphas and betas are also not able to travel long distances. Even more, they can easily be stopped by a thin wall, as shown in Figure 2.1.



Fig. 2.1 Penetration of alpha, beta and gamma radiation[8].



Fig. 2.2 Exposures from a radioactive cloud existing of (noble) gasses and aerosols[3].

2.2 How do radioactive materials disperse?

During a nuclear accident, the released materials are divided into two groups. One group are noble gasses, which are materials which dilute easily in the air. The second group contains aerosols, radioactive isotopes which travel through the air like a radioactive plume following the wind[9].

Those aerosols emit radiation that could reach the ground and its population. Exposure like this is a part of the external exposure of radiation. Since alpha and betas are stopped after a short range, this emission is mostly due to gamma radiation. The exposure only affects during the passage of the radioactive plume. The other part of external exposure is by radiation from radioactive materials settled on the ground, also called deposition. The effect of this exposure can last much longer and stops when all radioactive materials has decayed or have been removed from the ground.

Other exposures are internal by ingestion of radioactive food or inhalation. However, in this thesis only external exposures are included, since these can be sampled at measurement stations.

All exposures are shown in Figure 2.2. Besides the exposure paths, weather conditions play also an important role. An important weather condition is the mixing layer which is formed by discontinuities in temperature between upper and lower atmospheric layer which cause turbulence [10]. The mixing layer height, which varies during the day, affects the vertical diffusion of air pollutants and concentrations of water vapour.

2.3 How is radioactivity measured in the atmosphere?

Worldwide measurement networks as National Measurement network Radiation (NMR) in the Netherlands[11], detect radioactivity in the air and which are communicated through platforms as the EUropean Radiological Data Exchange Platform (EURDEP)[12]. The basic quantity of radioactivity is given by *activity*, the number of decayed nuclear particles, also called nuclides, per second, presented by the unit Becquerel (Bq)[3]. There are two types of gamma measurement stations. The first one measures the total gamma radiation per time. From this the specific materials can not be detected, however the measurement periods are quite small (a couple of minutes or hours) and increments of radioactivity can be detected. The other one measures fine dust in the air. It filters the air over a period of days and is able to detect different photon energies that are unique fingerprints for different materials.

Chapter 3

Problem description and previous work

3.1 Problem description

The problem that will be discussed in this thesis, is the reconstruction of a source term, here location, time and quantity of release, when only distant measurements of the release are known. This is a difficult problem since it depends highly on weather conditions, the used dispersion model and not fully reliable air samples. Mathematically this problem translates into the following inverse problem. If the actual dispersion can be represented by an operator H() the inverse problem is given by:

Solve
$$H(\sigma) = \mu$$
 for σ , (3.1)

where μ represents the measurements and σ the desired source term. Notice that this *H* operator may be very complex. Furthermore, because of discretely spaced measurement stations, multiple source terms may result in the same measurements. Therefore, the *H* operator generally does not have an inverse. For this reason this problem is often approached by a minimization problem of a previously chosen distance operator (*D*()) between the expected measurements from the model with the real measurements:

$$\min_{\sigma} D(H(\sigma) - \mu). \tag{3.2}$$

3.2 Previous work

Inverse problems have been noticed in many different fields as imaging and dispersion. Because of that, a lot of literature can be found on this topic and some of them are combined



Fig. 3.1 Example of Backtrack trajectories from Leipzig at heights 4200 m (red), 4600 m (blue) and 5200 m (green)[16].

and summarized in works as Rao (2007) and Hutchinson (2017) [13, 14]. Some of those methods for solving inverse problems are discussed in this section.

Backtrack trajectories and trial-and-error

The easiest and most intuitive way of solving such an inverse problem is by using that previous wind fields are known and have a great influence on the dispersion of the pollutants. According to this idea, one could think of calculating trajectories back in time from the measurement stations with only respect to the wind fields at that moment[15]. It is intuitive to think that all trajectories will cross each other in the neighborhood of the actual release location. However the dispersion depends on not only the weather, but also decay and deposition. Since some important information are missing, the solution area, as well in time and quantity of release, would therefore be very large. Hence many scientists chose to find the best source term by trail-and-error. This method corresponds with trying multiple possible source terms, generated by the backward trajectories, to minimize the distance between modeled measurements, obtained by a simulated release from the possible source term, and real measurements.



Fig. 3.2 Left: Ground concentration at arrival time of the cloud for a simulated release from Edinburgh with the corresponding sampling locations represented by red stars. Right: The corresponding dual concentration fields (retro plumes) released from the sampling stations integrated over time.

Dual dispersion model

A more advanced way of backtracking is by a dual dispersion model proposed by Pudykiewicz (1998) [17]. Theoretically, this is based on the adjoint operator constructed from a linear operator representing the forward dispersion model ($H(\sigma)$). In the following section a dual dispersion model will be derived and discussed in more detail.

A nice property of the dual model, is that the dual concentration field over time (*t*) and space (\vec{r}) for one measurement (*m*), also called retro plume, represents how much of the real released quantity would have been detected if the source term was at location \vec{r} at moment *t*. So dual concentrations are actually a representation of the sensitivity of a sampling station to a potential release at each location in time. Because of this, the forward operator $H(\sigma)$ can be approximated by a matrix *G* existing of dual concentrations in time and space for each measurement station, such that the measured quantities are approximately given by: $G\sigma \approx \mu$ for the actual quantity of release vector σ over time, location and radionuclides. The advantage of this matrix is that it can be computed in advance of the reconstruction because only the moment of measurement and measurement locations are used and not the exact measured quantities yet. As a result, many scientists worked on the ill-posed minimization of $D(\mu - G\sigma)$. It is ill-posed since *G* is in general a huge matrix and not necessarily a squared matrix.

One way of solving the linear system $G\sigma = \mu$ for σ is by variational methods. Such methods minimize a previous formulated cost function based for instance on a maximum or relative entropy with the prior being for instance a Bernoulli law or Poisson-like law [18, 19], or a

least squared function with a Tikhonov regularization [20]. In these cases, prior knowledge is used to correct for the ill-posedness of the system. Prior knowledge can be for instance a most likely ratio between two radionuclides. Another method developed for source term reconstruction is based on Bayesian Monte Carlo, proposed by Yee et al (2008)[21]. With the theorem of Bayes a probability density function of a source term generating the given data can be approximated. This is done by a Gaussian representation of a plume dispersion model and comparison of measurements with expected measurements obtained by a simulation of possible source terms. Since the source term consists of many variables, such as location, moment and quantity of multiple radionuclides, the probability density becomes complex and therefore is often approximated by an Monte Carlo method.

Since the dual dispersion model is widely used for source reconstruction methods, for example by Krysta (2017)[18] and Yee (2018)[21], the aim of this project will be firstly to determine the most likely source term by a simple method with dual concentrations. This method will be based on a recent paper from Efthimiou (2017)[22] about a simple reconstruction method with correlations between measurements and dual concentrations. Secondly, the dual concentration and reconstruction method will be analyzed for the positioning of sampling stations, incomplete weather information and measurement and model errors.

Chapter 4

Method

4.1 Dispersion model

The forward dispersion model used for this project is a so called Puff-model provided by the RIVM[23]. This model simulates air pollutants as a small cloud with a Gaussian distribution in space. Such a small cloud moves forward in time according to the wind at its center and diffuses. When the small cloud is large enough, it splits itself into multiple clouds which move independently of each other. During this process radionuclides also decay. The speed of decay depends on the specific materials, for example Iodine-131 has a half live of 8.02 days[24], and Cesium-137 a half life of 30.15 years [25]. Nuclides also set down as wet deposition (for instance by rain) or dry deposition (heavy solid nuclear materials falling on the ground). This model is similar to an Eulerian advection-diffusion dispersion model on the domain $\Omega \subset \mathbb{R}^3$ over time interval [0, T] given by [17]:

$$L\phi = \frac{\partial\phi}{\partial t} + \nabla \cdot \mathbf{u}\phi - \nabla \cdot \mathbf{K}\nabla\phi + \lambda\phi = f, \qquad (4.1)$$

with *L* the linear operator representing the dispersion model and ϕ represents the activity of a radio nuclide per unit volume in the air. Furthermore, *f* is the forcing term, in this case it has a positive value at the release location and is zero everywhere else. The vector $\mathbf{u} \equiv (u_1, u_2, u_3)^T$ represents the wind field in three dimensions and $\nabla \cdot \mathbf{K} \nabla$ gives the diffusion where ∇ is the gradient, $\nabla \cdot$ the divergence and *K* a diagonal matrix with diffusion coefficients in each direction. Diffusion coefficients are proportionality constants between the diffusion in each direction and the gradient of the concentration, such that *K* becomes a diagonal matrix. The final λ represents the deposition and decay parameter for a radio nuclide [17]. To solve the system 4.1, the following initial and boundary conditions should be satisfied. At the first moment of the model, the concentration field is assumed to be known. For simplicity, this field can be zero everywhere away from the source. The time interval [0, T] over which the dispersion model is computed, is assumed to be large enough such that the concentration field is periodic with T. This means that at the final moment T the concentration field is the space domain and the materials have decayed, such that the concentration field is the same as before the release started. Furthermore, it is assumed that the simulated release is the only source of radioactivity and the computed area is large enough, such that no concentration goes out of the space domain and by that mass is conserved. Close to the boundaries an outflow of concentrations is assumed to be constant and near the ground the concentrations are assumed to decrease due to loss of concentrations are also assumed to be constant. Details of initial and boundary conditions can be found in Pudykiewicz (1998) [17].

It is very important that since the puff-model consists of small Gaussian distributed clouds, air pollutant concentrations at a moment in time can be computed individually for each location and independent of other clouds. Because of this, mass conservation in this model is met by the individual clouds. As a result of this, no boundary conditions in space, besides the ground, are required.

4.2 **Dual dispersion model**

In order to find the dual model of dispersion model 4.1, we have to determine the adjoint operator of L. First of all, let us start with the following definition of an adjoint operator in Hilbert space from Sobolev [26]:

Definition 1 (Adjoint operator in Hilbert space) Let $L : H_1 \to H_2$ be a linear operator between Hilbert spaces. Then the adjoint operator $L^* : H_2 \to H_1$ satisfies:

$$\langle Lx, y \rangle_{H_2} = \langle x, L^*y \rangle_{H_1}, \text{ for all } x \in H_1, y \in H_2,$$

$$(4.2)$$

with \langle , \rangle_{H_i} inner product in Hilbert space H_i .

With this definition many different adjoint operators can be found for our model, since it highly depends on the choice of boundary conditions for the dual dispersion model. However, with a particular choice of boundary conditions, the dual dispersion model in theorem 1 can be found. In this theorem p represents a forcing term of the dual dispersion model. This particular adjoint operator was proposed by Pudykiewicz (1998), a partial derivation of this

operator will be discussed here. The paper of Pudykiewicz (1998) [17] provides a more detailed derivation.

Theorem 1 The following dispersion model is, under the right conditions, an adjoint model of 4.1

$$L^*\phi^* = -\frac{\partial\phi^*}{\partial t} - \nabla \cdot \mathbf{u}\phi^* - \nabla \cdot \mathbf{K}\nabla\phi^* + \lambda\phi^* = p, \qquad (4.3)$$

with $\phi^* \in L_2(\Omega \times (0,T))$ and periodic with period T.

In order to prove theorem 1 the following lemma is showed to be true.

Lemma 1 The adjoint operator of

$$\mathbf{L} = \frac{\partial}{\partial t} + \nabla \cdot \mathbf{u} - \nabla \cdot \mathbf{K} \nabla + \lambda, \quad \mathbf{L} : H \to H,$$
(4.4)

with $H = L_2(\Omega \times (0,T))$, can be represented by the linear operator

$$\mathbf{L}^* = -\frac{\partial}{\partial t} - \nabla \cdot \mathbf{u} - \nabla \cdot \mathbf{K} \nabla + \lambda + F, \quad \mathbf{L}^* : H \to H,$$
(4.5)

with F existing of additional term due to integration by parts and Greens Theorem.

At first notice that the system 4.1 has the linear operator

$$\mathbf{L} = \frac{\partial}{\partial t} + \nabla \cdot \mathbf{u} - \nabla \cdot \mathbf{K} \nabla + \boldsymbol{\lambda},$$

with domain $\mathscr{D}(\mathbf{L})$ a subset of the Hilbert space $H \equiv L_2(\Omega \times (0,T))$ which are periodic in time with period *T*. In this way the Hilbert spaces in the definition of adjoint operator in Hilbert space are the same, such that:

$$\langle \mathbf{L}\boldsymbol{\alpha},\boldsymbol{\beta}\rangle = \langle \boldsymbol{\alpha},\mathbf{L}^*\boldsymbol{\beta}\rangle, \quad \boldsymbol{\alpha}\in\mathscr{D}(\mathbf{L}),\boldsymbol{\beta}\in\mathscr{D}(\mathbf{L}^*),$$
(4.6)

for the adjoint operator L^* . Recall that the inner product in L_2 is defined by:

$$\langle \gamma, \beta \rangle = \int_0^T \int_\Omega \gamma \beta d\Omega dt$$
, where $\gamma, \beta \in H$. (4.7)

Similar to the equation 4.1 for the dispersion operator, the adjoint operator can also be formulated in the form:

$$\mathbf{L}^* \boldsymbol{\phi}^* = p, \quad \boldsymbol{\phi}^* \in \mathscr{D}(\mathbf{L}^*), \tag{4.8}$$

with *p* a forcing term representing the measurements. Let $\phi \in \mathscr{D}(\mathbf{L})$ and $\phi^* \in \mathscr{D}(\mathbf{L}^*)$. With Greens Theorem and Integration by parts the following inner product can be written as:

$$\begin{split} \langle \mathbf{L}\phi, \phi^* \rangle &\equiv \int_0^T \int_{\Omega} \frac{\partial \phi}{\partial t} \phi^* + \nabla \cdot \mathbf{u}\phi \phi^* - \nabla \cdot \mathbf{K}\nabla \phi \phi^* + \lambda \phi \phi^* d\Omega dt \\ &= \int_0^T \int_{\Omega} -\phi \frac{\partial \phi^*}{\partial t} - \phi \nabla \cdot \mathbf{u}\phi^* - \phi \nabla \cdot \mathbf{K}\nabla \phi^* + \phi \lambda \phi^* d\Omega dt + \mathscr{F}_{\tau} + \mathscr{F}_{\partial\Omega} \\ &= \langle \phi, \mathbf{L}^* \phi^* \rangle, \end{split}$$

with

$$\mathscr{F}_{\tau} = \int_{\Omega} \phi^* \phi \big|_{t=0}^{t=T} d\Omega \text{ and}$$
$$\mathscr{F}_{\partial\Omega} = \int_0^T \int_{\partial\Omega} \phi^* \phi \mathbf{u} - \phi^* \mathbf{K} \nabla \phi + \phi \mathbf{K} \nabla \phi^* d\omega dt.$$

In this derivation \mathscr{F}_{τ} contains the remaining part after partial integration of the first term $\int_0^T \int_\Omega \frac{\partial \phi}{\partial t} \phi^* d\Omega dt$ and $\mathscr{F}_{\partial\Omega}$ contains the remaining parts of the other terms.

This proves that Equation 4.5 is indeed an adjoint operator of the original linear operator of the dispersion model. With a particular choice of boundary conditions for the dual dispersion model, we find that \mathscr{F}_{τ} and $\mathscr{F}_{\partial\Omega}$ becomes zero.

Lemma 2 With the following assumptions and boundary conditions \mathscr{F}_{τ} and $\mathscr{F}_{\partial\Omega}$ are zero:

- 1. $\vec{\phi}(\vec{r},t)$ and $\vec{\phi}^*(\vec{r},t)$ are periodic with period T,
- 2. no wind at the ground and in the highest air layer of the space domain,
- 3. there is no inflow of dual concentration, since all releases are within the space domain,
- 4. Al dual concentration stays within the space domain, since the it is chosen large enough,
- 5. near the ground dual concentration in the cloud decreases due to deposition,
- 6. in the highest air layer dual concentration is constant.

For the mathematical representation and correctness of this lemma, see the paper of Pudykiewicz (1998)[17].

Finally Lemma 1 and Lemma 2 together prove Theorem 1. This theorem provides us with a dual dispersion model to compute the sensitivity of measurement stations to possible source terms.

Remark that the current puff model only has a boundary in space near the ground. Through mass is conserved by the individual small clouds. Because of that, Items 3, 4 and 6 of lemma 2 do not have to be met according to the used model.

Furthermore, notice that the adjoint model in Theorem 1 is actual the original model 4.1 with reversed time and wind. The forcing term p can be chosen such that it represents the measurements, which provides us with a retro plume shown in Figure 3.2.

4.2.1 Properties of the dual dispersion model

Let us assume for simplicity that the release was instantaneous at one location, then there is a nice relationship between the real concentration, dual concentration and quantity released from the exact source term. The result of Theorem 1 is given by the following Corollary.

Corollary 1 If the real release was instantaneous at location $\vec{r_0}$, time t_0 and with a release of Q,

$$f(\vec{r},t) = \delta(\vec{r} - \vec{r_0})\delta(t - t_0)Q.$$
(4.9)

The corresponding retro plumes are released from measurement locations $\vec{r_m}$ at time t_m for m = 1...M with quantity 1/M

$$p(\vec{r},t) = \sum_{m=1}^{M} \frac{1}{M} \delta(\vec{r} - \vec{r_m}) \delta(t - t_m).$$
(4.10)

Then the following holds:

$$Q\phi^*(\vec{r_0}, t_0) = \sum_{m=1}^M \frac{1}{M}\phi(\vec{r_m}, t_m).$$
(4.11)

Here, $\delta()$ is the Dirac delta function. In words this means that the dual concentration at the exact source location and time times the exact amount of release, is equal to the sum of the modelled measurements at sampling stations.

Since no literature was found about the derivation of this result, although it is used through multiple papers by for example Krysta (2017)[18], Saunier (2013),[20], Yee (2018)[21] and Efthimiou (2017)[22]. We will prove and discuss this result in detail for different scenario's such as instantaneous measurements and averaged measurements.

In order to show the correctness of Corollary 1 first let f and p be as in this Corollary. By Theorem 1 we know that $\mathbf{L}\phi = f$ and $\mathbf{L}^*\phi^* = p$, such that the inner product between f and the dual concentration field is given by:

$$\langle f, \phi^* \rangle = \int_0^T \int_\Omega f(\vec{r}, t) \phi^*(\vec{r}, t) d\Omega dt = Q \phi^*(\vec{r_0}, t_0).$$

Furthermore, the in-product between p and real concentration field is calculated with:

$$\langle \phi, p \rangle = \int_0^T \int_\Omega p(\vec{r}, t) \phi(\vec{r}, t) d\Omega dt = \sum_{m=1}^M \frac{1}{M} \phi(\vec{r_m}, t_m).$$

Finally, the definition of an adjoint operator provides us with the following equality:

$$\langle f, \phi^* \rangle = \langle L\phi, \phi^* \rangle = \langle \phi, L^*\phi^* \rangle = \langle \phi, p \rangle,$$

which implies that Corollary 1 is true.

Besides, the forcing term p is a summation over measurements. Since p influences the calculations of the retro plumes, one can write the dual concentration as a summation over dual concentrations for each measurement station:

$$\phi^*(\vec{r},t) = \sum_{m=1}^M \frac{1}{M} \phi_m^*(\vec{r},t).$$

This means that Corollary 1 is also true for each measurement station because of the freedom of choosing those stations.

Corollary 2 For an instantaneous release from the actual source location and instantaneous releases with the dual model from the measurement stations, even

$$Q\phi_m^*(\vec{r_0}, t_0) = \phi(\vec{r_m}, t_m) \tag{4.12}$$

holds for every measurement m.

Until now, measurements were assumed to be instantaneous. From now on assume that only averaged concentration over a known period are known, which is true for real measurements.

In this case Corollary 3 states that the same relation between the dual and forward concentration fields still holds.

Corollary 3 Equation 4.12 holds also if the concentrations of the real release are averaged over time, here denoted by the period $[t_m, t_m + n_m dt]$ for each measurement m over n_m time

steps of dt. Than the dual force term is:

$$p(\vec{r},t) = \sum_{m=1}^{M} \frac{1}{Mn_m} \int_{t_m}^{t_m + n_m dt} \delta(\vec{r} - \vec{r_m}) \delta(t - \tilde{t}) d\tilde{t}.$$
 (4.13)

With p as in Corollary 3 the dual concentration is also averaged over time and the inner product between the forward concentration and p becomes:

$$\langle \phi, p \rangle = \sum_{m=1}^{M} \frac{1}{Mn_m} \int_{t_m}^{t_m + n_m dt} \phi(\vec{r_m}, \tilde{t}) d\tilde{t} = \sum_{m=1}^{M} \frac{1}{M} \tilde{\phi}(\vec{r_m}),$$

with $\tilde{\phi}(\vec{r}_m)$ the forward concentration averaged over $[t_m, t_m + n_m dt]$. In that case equation 4.12 still holds:

$$Q\phi_m^*(\vec{r_0},t_0) = \tilde{\phi}(\vec{r}_m).$$

With respect to retro plumes, the dual concentration is now the average value over n_m unitary releases from the location of measurement *m* at the sampling interval.

Unfortunately, in many cases the exact concentrations at each time are not known and the only knowledge of air pollutants is given by measurements measured over some period of time. So assume that one measurement is given by:

$$\mu_m = \tilde{\phi}(\vec{r}_m) + \varepsilon_m^{meas},$$

with measurement error ε_m^{meas} . The influence of measurement errors on the computation of Q for each measurement can be determined as follows. The dual concentration is independent of measurement error, so the expected quantity \hat{Q} would be given by:

$$\hat{Q} = \frac{\mu_m}{\phi_m^*} = \frac{\tilde{\phi} + \varepsilon_m^{meas}}{\phi_m^*} = Q + \frac{\varepsilon_m^{meas}}{\phi_m^*}.$$
(4.14)

Since dual concentrations are from a unitary release, they are considerably small. Because of that, measurement errors highly increase errors in the computation of quantity of release. The smaller the dual concentration, the higher the expected quantity of release becomes and more sensitive \hat{Q} for measurement errors. As a result of measurement errors, sampling stations far away from the actual source shows more uncertainty in the reconstruction of the amount of release than stations close to the source.

Actually, a release happens in general over a time interval of multiple hours, for example during a nuclear accident at a power plant. In such a case, the result given by Equation 4.12

does not hold any more. This is because the forcing term f of the dispersion model should now be integrated over a time period. By that, the dual concentrations and computed quantity of release are also integrated as shown in Corollary 4.

Corollary 4 If the release was over time interval $[t_0, t_1]$, the forcing term of the dispersion model becomes

$$f(\vec{r},t) = \int_{t_0}^{t_1} \delta(\vec{r} - \vec{r_0}) \delta(t - \hat{t}) Q(\hat{t}) d\hat{t}$$
(4.15)

and the following holds:

$$\int_{t_0}^{t_1} Q(t)\phi_m^*(\vec{r_0}, t)dt = \phi(\vec{r_m}, t_m), \qquad (4.16)$$

for each measurement m.

4.3 Method description

Efthimiou (2017) [22] proposed a source term reconstruction method based on correlations. Because of corollary 3 ($\mu = Q\phi^*$) the dual concentrations and measurements are expected to correlate at the real release location and time of release. From this relation, Efthimiou formulated the following correlation based cost function:

$$J = -\frac{\langle (\mu - \langle \mu \rangle)(Q\phi^* - \langle Q\phi^* \rangle) \rangle}{\sqrt{\langle (\mu - \langle \mu \rangle)^2 \rangle} \sqrt{\langle (Q\phi^* - \langle Q\phi^* \rangle)^2 \rangle}},$$
(4.17)

where $\langle \rangle$ denotes arithmetic averaging over all measurements. The best solution would than be found by a minimization of cost function *J*. Recall that this cost function does not depend on Q, since a correlation does not depend on a multiplication with an constant.

However, after some tests, it became clear that an high correlation does not always give the best solution. Recall that a correlation between vectors $\vec{\mu}$ and $\vec{\phi}^*$ represents the fit of data on the linear form: $\mu = a\phi^* + b$. However, we know for the actual source term, that *b* should be zero and *a* equal to the quantity of release *Q*. According to tests we found that the value of *b* becomes more negative further from the source such that the correlations becomes higher than expected. This results in more uncertainty in the source term reconstruction. Correlations and the corresponding values of *b* are shown in Figure 4.1. In order to correct for overestimated correlations, a method is proposed where correlation is computed for a linear regression through the origin, so where the average value of the expected and real measurements are equal.

Let $\vec{\mu}$ and $\vec{\phi}^*(\vec{r},t)$ be vectors of length *M* with elements of respectively the measurement and dual concentration for each of the *M* measurements. Than the correlation, forced trough



Fig. 4.1 Maximal correlations (light blue shadow) and corresponding y-intersect (pink shadow), the value of b in $\mu = a\phi^* + b$ with unit Bq/m³, according to method of Efthimiou for a release from Edinburgh with a South-East wind.

the origin, for a possible release location \vec{r} at moment *t* can be found by:

$$C(\vec{r},t) = Corr_0(\vec{\mu}, \vec{\phi^*}(\vec{r},t)) = \frac{\vec{\mu}^T \vec{\phi^*}}{\sqrt{(\vec{\phi^*})^T \vec{\phi^*}(\vec{\mu})^T \vec{\mu}}}.$$
(4.18)

Notice that the slope of the linear regression would give the expected amount of released materials. Q can than be estimated by:

$$\hat{Q}_{\vec{r},t} = \frac{\vec{\mu}^T \vec{\phi}^*}{(\vec{\phi}^*)^T \vec{\phi}^*}.$$
(4.19)

Let the set of all possible source locations be *PSL*. In order to find the most likely source location, the time for each possible source $p \in PSL$ at location \vec{r}_p is chosen to be the moment which maximizes the correlation over time:

$$t_p = argmax_t \{ C(\vec{r}_p, t) \}.$$

$$(4.20)$$

In this way the solutions are reduced to the number of possible source locations and corresponding computed released quantity is denoted by \hat{Q}_p .

To score each location we use the Root Mean Squared Error, defined by $RMSE(x) = \sqrt{\frac{x^4x}{N}}$ for an error vector $x \in \mathbb{R}^N$. In this way the score of possible source location p is computed by:

$$R(p) = RMSE(\vec{\mu} - \hat{Q}_p \vec{\phi}^*(\vec{r}_p, t_p)).$$

$$(4.21)$$

We found a nice effect of the Root Mean Squared Error in case of an instantaneous release. Then the RMSE can be written as a function of correlation and measurements. This function is derived below:

$$\begin{split} \|\vec{\mu} - \hat{Q}_{p}\vec{\phi}^{*}(\vec{r}_{p}, t_{p})\|_{2}^{2} &= \vec{\mu}^{T}\vec{\mu} - 2\hat{Q}_{p}\vec{\mu}^{T}\vec{\phi}^{*} + \hat{Q}_{p}^{2}\vec{\phi}^{*T}\vec{\phi}^{*} \\ &= \vec{\mu}^{T}\vec{\mu} + \hat{Q}_{p}(-2\vec{\mu}^{T}\vec{\phi}^{*} + \hat{Q}_{p}\vec{\phi}^{*T}\vec{\phi}^{*}) \\ &= \vec{\mu}^{T}\vec{\mu} - \hat{Q}_{p}\vec{\mu}^{T}\vec{\phi}^{*} \\ &= \vec{\mu}^{T}\vec{\mu} - \frac{(\vec{\mu}^{T}\vec{\phi}^{*})^{2}}{(\vec{\phi}^{*})^{T}\vec{\phi}^{*}} \\ &= \|\vec{\mu}\|_{2}^{2} \left(1 - Corr_{0}^{2}(\vec{\mu}, \vec{\phi}^{*}(\vec{r}_{p}, t_{p}))\right), \end{split}$$

with $\|\cdot\|_2$ the l^2 - norm.

We now introduce the RMSE reduction as a fraction of RMSE without the method, so $RMSE(\vec{\mu})$, and RMSE with the method, so with the computed amount of release \hat{Q}_p (as was used in a source term reconstruction report from the IRSN [7]). The RMSE reduction is then shown by:

$$Red(p) = \frac{RMSE(\vec{\mu})}{RMSE(\vec{\mu} - \hat{Q}_p \vec{\phi}^*(\vec{r}_p, t_p))} = \frac{1}{\sqrt{1 - Corr_0^2(\vec{\mu}, \vec{\phi}^*(\vec{r}_p, t_p))}}.$$
(4.22)

The best source term is than calculated by the maximization of RMSE reductions. In case of instantaneous releases this method corresponds with a maximization of the correlations.

Previously, we assumed that the real release was instantaneous, however, this is in general not the case, as was discussed before. If a long release over time period T_0 is allowed the method becomes more complex, since the RMSE reduction becomes:

$$\frac{RMSE(\vec{\mu})}{RMSE(\vec{\mu} - \int_{T_0} \hat{Q}_p(t)\vec{\phi}^*(\vec{r}_p, t)dt)} = \frac{1}{\sqrt{1 - \int_{T_0} Corr_0^2(\vec{\mu}, \vec{\phi}^*(\vec{r}_p, t))dt}}.$$
(4.23)

The method than requires a continuous field of dual concentration. However, the used puff model only computes dual concentrations discrete in time and space. Because of that and to keep the method simple, we assume the release was instantaneous.

The proposed reconstruction method maximizes the Root Mean Squared Error reduction over possible source locations $p \in PSL$:

$$\max_{p} Red(p). \tag{4.24}$$

Since the actual release is assumed to be instantaneous the RMSE reduction is given by the following function of correlation between measurements $\vec{\mu}$ and dual concentrations $\vec{\phi}^*(\vec{r},t)$:

$$Red(p) = \frac{1}{\sqrt{1 - C^2(\vec{r}_p, t_p))}},$$

with
$$C(\vec{r},t) = \frac{\vec{\mu}^T \vec{\phi}^*(\vec{r},t)}{\sqrt{(\vec{\phi}^*(\vec{r},t))^T \vec{\phi}^*(\vec{r},t) \vec{\mu}^T \vec{\mu}}}, \quad t_p = argmax_t \{C(\vec{r}_p,t)\}$$

and $\hat{Q}_p = \frac{\vec{\mu}^T \vec{\phi}^*(\vec{r}_p,t_p)}{(\vec{\phi}^*(\vec{r}_p,t_p))^T \vec{\phi}^*(\vec{r}_p,t_p)}.$

4.3.1 Measurement and model error dependency of the method

Since the only information about the release comes from sampling stations, it is important to know how the method reacts to measurement errors. For this, assume that a measurement μ_m can be represented by what should have been measured v plus some measurement error ε_m^{meas} :

 $\mu_m = v(\vec{r}_m, t_m) + \varepsilon_m^{meas} = Q\phi_m^* + \varepsilon_m^{meas}$, for every measurement *m*.

Then the computation of the quantity would show the following relation with the measurement errors:

$$\hat{Q} = \frac{\vec{\mu}^T \vec{\phi}^*}{(\vec{\phi}^*)^T \vec{\phi}^*} = \frac{(Q\phi^* + \varepsilon^{\vec{meas}})^T \vec{\phi}^*}{(\vec{\phi}^*)^T \vec{\phi}^*}$$
$$= Q + \frac{\varepsilon^{\vec{meas}}^T \vec{\phi}^*}{(\vec{\phi}^*)^T \vec{\phi}^*}$$

Since the dual concentrations are computed from a unitary release, $\vec{\phi}^*$ is very small, of order $\mathscr{O}(\frac{\max \vec{v}}{Q})$. Because $\varepsilon^{meas} \gg \phi^*$, the error ε^{meas} has a great influence on the computed quantity of release. However, according to this method the performance of a solution given

by the Root Mean Squared Error is independent of the computed measurements and highly depends on the correlation between the measurements and modelled dual concentrations. By a computation of the correlation through the origin, the influence of measurement errors can be computed as follows.

$$corr_{0}(\vec{\mu},\vec{\phi}^{*}) = \frac{\vec{\mu}^{T}\vec{\phi}^{*}}{\sqrt{(\vec{\phi}^{*})^{T}\vec{\phi}^{*}(\vec{\mu})^{T}\vec{\mu}}} = \frac{\frac{(\vec{\nu}+\epsilon^{\vec{m}eas})^{T}\vec{\phi}^{*}}{\sqrt{(\vec{\phi}^{*})^{T}\vec{\phi}^{*}}}}{\sqrt{(\vec{\mu})^{T}\vec{\mu}}}$$
$$= \frac{corr_{0}(\vec{\nu},\vec{\phi}^{*})\sqrt{(\vec{\nu})^{T}\vec{\nu}} + corr_{0}(\epsilon^{\vec{m}eas},\vec{\phi}^{*})\sqrt{(\epsilon^{\vec{m}eas})^{T}\epsilon^{\vec{m}eas}}}{\sqrt{(\vec{\mu})^{T}\vec{\mu}}}$$
$$= \frac{\sqrt{(\vec{\nu})^{T}\vec{\nu}} + corr_{0}(\epsilon^{\vec{m}eas},\vec{\phi}^{*})\sqrt{(\epsilon^{\vec{m}eas})^{T}\epsilon^{\vec{m}eas}}}{\sqrt{(\vec{\mu})^{T}\vec{\mu}}}$$

Here it can be seen that correlation highly depends on the correlation between the measurement error and the dual concentration. Since we expect a high correlation between dual concentration and measurements, this means that ideally measurement errors are independent of measured quantities at sampling stations.

Furthermore, besides the measurements, the model also has errors in the computation of the dual concentrations. Assume that the modelled dual concentration $\vec{\phi}^*$ consist of the sum of the real dual concentration $\vec{\phi}^*$ and the model error η . With measurement error ε^{meas} and model error multiplied by the quantity of release ε^{mod} , the measurements can be written as:

$$\mu_m = Q(\phi_m^* + \eta_m) + \varepsilon_m^{meas} = v(\vec{r}_m, t_m) + \varepsilon_m^{mod} + \varepsilon_m^{meas}, \text{ for every measurement } m$$

With both model and measurement errors we find the following changes in computation of the quantity of release and correlation:

$$\hat{Q} = Q + \frac{\varepsilon_{mod}^{T}\vec{\phi}^*}{(\vec{\phi}^*)^T\vec{\phi}^*} + \frac{\varepsilon_{meas}^{T}\vec{\phi}^*}{(\vec{\phi}^*)^T\vec{\phi}^*},$$

$$corr_{0}(\vec{\mu},\vec{\phi}^{*}) = \frac{\sqrt{(\vec{\nu})^{T}\vec{\nu}} + corr_{0}(\vec{\varepsilon}_{mod},\vec{\phi}^{*})\sqrt{(\vec{\varepsilon}_{mod})^{T}\vec{\varepsilon}_{mod}} + corr_{0}(\vec{\varepsilon}_{meas},\vec{\phi}^{*})\sqrt{(\vec{\varepsilon}_{meas})^{T}\vec{\varepsilon}_{meas}}}{\sqrt{(\vec{\mu})^{T}\vec{\mu}}}$$

Although the model error η may be very small, the multiplication with Q results in a great influence of ε^{mod} on the computation of Q. We expect that the influence of the model error decreases over time, because of diffusion. Instantly after the retro plume is

released, the clouds of dual concentrations are strongly concentrated and because of that, the modeled concentration field is highly dependent on the discretized wind field. Due to the discretization of the wind field, plume trajectories can diverge from reality. However, this does not necessarily mean that dual concentrations are proportional with model errors, since the wind field can be accurate at the time of release.

4.3.2 Dependency of computed amount of release for choice of measurements

At first the measurements are given in Bq/m^3 and the quantity of release in Bq. Because of the equation $\mu = Q\phi^*$ (equation 4.19) the unit of dual concentration is per cubic metres (1/m³).

Furthermore, the method is strongly dependent on the choice of measurements. Assuming that the measurements lie on the trajectory of the plume centre of the actual release, the centre of retro plumes are expected to travel over the exact release location. Moreover, the puff model computes individual Gaussian distributed clouds, thereby concentrations of the actual release and retro plumes can be approximately represented by a bi-variate normal distribution. Let us assume that the clouds only diffuse in horizontal directions, since after a few hours a cloud is already mixed up in vertical direction under an air layer called the mixing layer. The decrease of a local concentration over time can be represented by the change of the highest point of a bi-variate normal distribution when the standard deviation increases with time. Assumed that the standard deviation increases linear with time according to $\sigma(t) = \alpha t + \beta$ and since only the centre of the plume is of interest, the dual concentration is expected to be approximately given by:

$$\phi_m^*(t) = \frac{1}{2\pi\sigma^2(t)} = \frac{1}{2\pi(\alpha t + \beta)^2}$$
(4.25)

$$\approx \frac{1}{2\pi\alpha^2} t^{-2} = \gamma t^{-2}, \qquad (4.26)$$

assuming that $\beta \ll \alpha t$ for a standard deviation varying over time by $\sigma(t) = \alpha t + \beta$ and $\gamma \equiv \frac{1}{2\pi\alpha^2}$. However, the first assumption about measuring the centre of the plume released with the actual source term, is a really strong assumption. If the measurement was at the edge of the plume, the sampled concentration would be far lower than along the centre trajectory. This means that in practice γ would be much smaller than determined in this way. Since many measurements are not along the centre of the plume, an expected value of dual concentrations are difficult to obtain.

When we take a look at the computed quantity it is noticeable it depends highly on an inner product between measurements and dual concentrations. According to this, if a subset of the measurements with low measured quantities are chosen, the method would probably give an underestimation of the probable quantity of release. The factor between different quantity computations of a varying set of measurements is unfortunately hard to obtain, since dual concentrations are difficult to approximate.

Furthermore, a possible release location between the sample stations and the real release location is expected to show a lower possible release amount than was actually released. Similarly, a larger release is expected further away from the measurement stations. In order to predict how the amount of release progresses over time, we use the fact that in any case the product of dual concentration with the amount of release should be the measured concentration $(Q\phi^* = \mu)$. Let Q at a possible source location be q_{psl} and at the actual source location Q_{Source} . With help of the approximation of dual concentration by Equation 4.26 the expected quantity at location psl and time t_{psl} for one measurement m can be calculated by the ratio of expected dual concentrations.

$$q_{psl} = Q_{Source} \frac{\phi_{Source}^*}{\phi_{psl}^*} = Q_{Source} \frac{(dt_m + dt_{psl})^2}{dt_m^2},$$

with dt_m the time to travel from the measurement station to the real source location and dt_{psl} the time from the source location to the *psl* location. For multiple measurements, the combined dual concentration is the average of individual unitary releases, so the factor is averaged over measurement stations.

$$q_{PSL} = Q_{Source} \sum_{m=1}^{M} \frac{1}{M} \frac{(dt_m + dt_{psl})^2}{dt_m^2}$$
(4.27)

Notice that Equation 4.27 can be rewritten as a polynomial of two degrees, depending on the choice of measurements.

$$q_{PSL} = Q_{Source} \sum_{m=1}^{M} \frac{1}{M} \frac{(dt_m + dt_{psl})^2}{dt_m^2}$$

= $Q_{Source} \sum_{m=1}^{M} \frac{1}{M} (1 + \frac{dt_{psl}}{dt_m})^2$
= $Q_{Source} \left[\left(\sum_{m=1}^{M} \frac{1}{M} \frac{1}{dt_m^2} \right) dt_{psl}^2 + 2 \left(\sum_{m=1}^{M} \frac{1}{M} \frac{1}{dt_m} \right) dt_{psl} + 1 \right]$
Chapter 5

Results

In this chapter the performance of the proposed reconstruction method described in Section 4.3 will be discussed according to three test cases. The goal of the first two cases is to test the performance of the method for station positioning with two simple manual generated wind fields. Since measurements are the only knowledge of the release of radioactive materials, they highly influence the solutions of the source term reconstruction method. The used wind fields are generated such that the first wind field blows straight forward and the second rotates each time step, resulting in a spiral shaped plume. For the test cases with those wind fields measurements are obtained by simulated incidents. Furthermore, the second test case with a rotational wind field also give us the opportunity to test for incomplete weather information. The KNMI provides current weather information only at every 3 or 6 hours and the dispersion model does discretized calculations in time and space for each radioactive cloud. This is expected to result in computational errors. Thereby, for performance the proposed method is tested for incomplete weather information for the rotating wind case. Finally, with help of the two manual generated wind fields, the expected performance of the method for real-life cases is discussed and are compared with the method's performance on the real European Tracer Experiment ETEX1. Since the actual source term of ETEX1 is known, the influence of measurement and model error are discussed to capture the effect of differences between the real and simulated world.

5.1 How the method is tested

The method is based on retro plumes, which are simulations of unitary releases located at sampling stations and computed by using the dual dispersion model. To simulate those plumes, source terms have to be defined for each measurement. For this we assume each measurement station is located at ground level, which is around 10 meters above the earth's surface. No additional heat is released and the plume starts with standard deviation of 20 meters in horizontal and vertical directions. Tests of releases at different heights between 5 and 200 meters, showed that dual concentrations only slightly change for different release heights and the height of release hardly influences the time the cloud passed by possible source locations. Furthermore, since the released material is not always known, especially in the early stage of release, the released material is assumed to be Cesium-137, a substance with a half life of 30.15 years [25]. Because of the slow decay of Cesium, incidents from long before the moment the stations have measured increments, will still be possible to be reconstructed.

During the test cases, the effect of deposition is not included in the dispersion model for simulation of measurements and in the dual dispersion model for simulation of retro plumes. Depending on the released materials, the scale of the problem and rain during the time period of simulation, the effect of deposition might be necessary to be included in the dispersion models. The proposed method can relatively easy be extended to include this effect.

As could be expect from the method description, this method requires possible source locations to compute the RMSE reductions in this area. To achieve this a Possible Source Location (PSL) grid is made in advance, based on a simulation of the joined retro plumes of each measurement station. According to this, can be seen which part of the earth will be covered by the clouds of dual concentrations and could be a possible source location. The required size and number of points in a PSL grid to obtain an accurate solution field, depends on the scale of the problem and the weather during this period. During retro plume simulations, dual concentrations are computed exactly at the locations of the PSL grid at every hour sharp for a grid of 0.25 degrees in latitude and longitude, which is approximately 17 by 28 kilometers. Relatively, this grid is chosen because it is a quite fine grid which can still be computed within a fine resolution in time.

The computational costs of the reconstruction method can be represented by the number of floating point operators (FLOPS). The number of floating point operators for the calculation of correlation and Q at each time and location, is approximately given by the number of psl's times the number of time steps and multiplied by ten times the number of measurements. The measurements are multiplied by ten, since the correlation and the computation of released quantity together contains five inner products, with each 2n - 1 FLOPS for vectors of length n, two divisions and a square root. According to this cost representation, each variable only influences the costs linearly. However, the number of psl's and measurements increases quadratic when the distances in each direction are reduced. In this way the additional costs of a finer measurement or PSL grid can be calculated.

In the following subsection a couple of parameters are introduced. Whereby the performance of the proposed source term reconstruction method on a straight wind field, a rotational wind field and ETEX1 can be determined.

5.1.1 Definitions of result representations

Solution fields computed by the method are represented by the solutions for which the reduction factors defined by Equation 4.21 exceeds factor f with a default value of f = 2. Furthermore, the uncertainty of the source term increases with the size of the solution field. To correct for the number of solutions the following distribution field is defined for the solution field of factor f:

Definition 2 (Location probability) A possible source location has the following probability of being the desired source term.

$$P_f(psl) = \frac{Red_f(psl)}{\int_{PSL_f} Red_f(s)ds}$$
(5.1)

where $Red_f(psl)$ is the maximal reduction at location $psl \in PSL_f = \{s \in PSL | Red(s) \ge f\}$.

All three test cases are chosen because the source terms are known. For the first two synthetic cases the measurements are simulated at a chosen source term and for the ETEX1 experiment the actual source term is known. Since all actual source terms are given, it is possible to sort the solutions from the solution field by performance compared to the actual source term. Herefore we define an expected progress of released quantity and distance to the actual source location depending on the time difference between the release time of possible solutions and of the actual source term.

First let us define a ratio between the actual and computed quantity of release by the following Q-factor.

Definition 3 (Qfactor) *The ratio between actual and calculated quantity of release is defined by:*

$$Qfactor(\hat{Q}, Q_0) = max\{\hat{Q}/Q_0, Q_0/\hat{Q}\}$$
 (5.2)

, with the actual release Q_0 and \hat{Q} computed by the method.

Assumed that the averaged wind speed is 5 m/s and the pollutants are already mixed completely in the vertical direction, we expect after one hour a travel distance of 18 km and by Equation 4.27 a Q-factor given by $\frac{1}{M}\sum_{m=1}^{M} \frac{(dt_m + dt_{PSL})^2}{dt_m^2}$. Since these computed Q-factors are really strict close to the actual source location, we use an acceptable factor of 4 and allow the quantity to increase two times as fast as we expected. In this way the expected Q-factor is given by

$$E_{Qfactor}(dt_{psl}) = \left(\sum_{m=1}^{M} \frac{1}{M} \frac{1}{dt_m^2}\right) (2dt_{psl})^2 + 2\left(\sum_{m=1}^{M} \frac{1}{M} \frac{1}{dt_m}\right) |2dt_{psl}| + 4, \quad (5.3)$$

in this calculation the absolute value of dt is used, such that the same Q-factor is expected at adjacent and following times with respect to the actual release time. Now, the expected distance and quantity of release with respect to the actual source term are given by functions depending on time. To be specific, on the difference in release time between the solutions and actual source term. Hereby every hour provides boundary values for time difference, distance and quantity difference with respect to the actual source term. The boundaries at each 6 hours are denoted as layer. Each solution is then sorted by those layers, such that the layer of a solution is the lowest layer for which all three variables of the solution are contained by the boundaries given by this layer. In this way layer 1 only contains possible solution with a source term within 108 kilometers of the source location, with a maximum time difference of ± 6 hour from the actual release time and a Qfactor below $E_{Qfactor}(6)$. For simplicity we define an accurate solution field by a field for which the majority of the solutions are within layer 4 or lower, this corresponds with a time accuracy of ± 24 hours.

In order to determine the density of measurement stations for each test case, measurement density is defined as below.

Definition 4 (Measurement density) *Measurement density is the number of stations per travel hour of the plume released from the real source location.*

Measurement density =
$$\frac{M}{t_{max} - t_0}$$
 (5.4)

, with *M* the number of measurements used in the method, t_{max} the last time of sampling and t_0 the actual release time.

Notice that this definition is only usable when the exact source term is known. If this is not the case the first sampling time can be used instead of the moment of release. However, this increases the measurement density considerably if the measurements are far away from the actual source location.

Finally, we also define a parameter which represents the confidence of the computed quantity of release based on the variation of the Q-factor according to the mean of the interval.

Definition 5 (Variation of quantity of release (Q)) Let a quantity interval of the set $\{q_1, ..., q_n\}$ be obtained at the time interval of length n for which the RMSE reduction exceeds factor f and q_0 and q_{n+1} respectively the adjacent and following quantities in time. Variation of quantity of release is than calculated by:

$$\sqrt{\frac{\sum_{i=0}^{n+1} Qfactor(q_i, \overline{Q})^2}{n+2}}, \text{ with } \overline{Q} = \frac{1}{n} \sum_{i=1}^n q_i.$$
(5.5)

In words, it is a variation of the Qfactor with an adjacent and following quantity in time with respect to the mean of the given quantity interval.

5.2 Synthetic cases

In this section the performance of the proposed reconstruction method is tested with two synthetic cases, in particular a straight wind field and a rotational wind field. The goal is to determine its performance for basic changes as different densities of measurements, a straight wind and rotational wind field, incomplete meteorological data and a large time gap between first measurement and the actual moment of release. For those synthetic cases measurements are modelled by ground concentrations averaged over one hour for a instantaneous simulated release of 10 minutes. The synthetic measurements are assumed to be the first averaged ground concentrations above zero. Furthermore, wind fields are manually generated where wind direction and speed may differ over time, though is the same for each location and height in the space domain. The mixing layer is always assumed to be constant at 700 meters height.

5.2.1 Straight wind

The simplest wind field is when the wind blows with a constant speed in one direction. In this test case an instantaneous release from Edinburgh is simulated with a constant wind speed of 7 m/s in South East direction over 7 days. The corrsponding simulated plume can be seen in Figure 5.1. The exact source term is given by a release from Edinburgh at the 5th of July at 9:00 AM with a released quantity of 1E+16 Bq Cesium-137.

The proposed method is tested for different densities of measurement stations. The measurement stations are positioned on a grid with equal distance in longitude and latitude degrees, such that in latitude the measurement stations are around respectively 400, 300, 200 and 100 km distance of each other. However, notice that the longitude distance in kilometers



Hourly integrated ground concentration at arrival time for a simulated release from Edinburgh

Fig. 5.1 Straight wind case. Left: concentrations integrated over 1 hour at arrival time of the cloud to simulate measurements of a release from Edinburgh. Right: time integrated retro plumes for measurement grid 2.

is smaller than the latitude distance. The specifics about these grids and there densities are shown in Table 5.1.

Grid Nr.	dx (lon- and	$dlon \times dlat (km)$	Nr. of	meas. density
	lat degrees)		stations	(stations/travel hour)
1	3.5	240×392	14	0.11
2	2.625	180 imes 294	27	0.19
3	1.75	120×196	59	0.38
4	0.875	60 imes 98	234	1.48

Table 5.1 Details about the measurement stations grids in case of a straight wind field.

The actual positions of the measurement stations and the measured quantities are shown in figure A.1 in Appendix A.

Measurement stations from grid 2 provides us with retro plumes shown in Figure 5.1. From such retro plumes the correlations and corresponding RMSE reductions are obtained. Examples of correlations and RMSE reduction over time for two locations are shown in Figure A.2 in Appendix A. Because of the relation given by Equation 4.22 between these two parameters, the RMSE reduction increases hyperbolic with correlations and by that shows only a peak exceeding 2.0 if the correlation is above $\frac{1}{2}\sqrt{3} \approx 0.866$.

The method provides us with a solution field given by the possible source locations which exceeds a RMSE reduction of 2.0. The solution fields for all four measurement grids are

Grid	PSL's	Dist (km)	Time interval	Quantity interval (Bq)	Red. interval
1	733	1965	[3 July 00:00, 6 July 00:00]	[1.65E+13, 5.06E+15]	[2.0, 9.7]
2	477	1740	[3 July 00:00, 6 July 00:00]	[2.65E+13, 6.06E+15]	[2.0, 8.2]
3	171	1111	[3 July 21:00, 5 July 20:00]	[8.43E+13, 7.71E+15]	[2.0, 14.2]
4	40	483	[4 July 17:00, 5 July 13:00]	[7.97E+14, 4.96E+16]	[2.0, 12.6]

Table 5.2 General results of the method with factor 2.0 for a straight wind for four different measurement grids. Grid 1: latitude distance of 400km between stations, grid 2: 300km, grid 3: 200km and grid 4: 100km. This table contains the number of possible source locations, the maximal distance between two possible source locations and total time, quantity and reduction intervals. With uncertainty in distance of ± 65 km and in time of ± 1 hour.

shown in Figure 5.2. The filling shows the probability $P_2(psl)$, defined by Equation 5.1, for each possible source location. Before we discuss these figures the general results are shown.

Although these figures can show the likely source term area, it does not say anything about the probable time and quantity of release. Therefore, in Table 5.2 the general results are shown. The second column shows the total number of possible source locations and the third column represents the area of the field. This is calculated by the largest distance between two edges of the largest rectangle which contains the whole solution field. The time and quantity intervals are given by the minimum and maximum values for time and Q. As expected, from Figure 5.2 and Table 5.2 can be noticed that a finer measurement grid gives a smaller possible source area and simultaneously a smaller time interval and higher location probabilities. However, it is remarkable that the actual quantity of release is only contained by our finest grid. The reason for this is the choice of different measurement locations and measured quantities. Since grid 1 has the fewest measurement stations, the chance of measuring high quantities is small, while a finer measurement grid as grid 4 has a much larger range of measured quantities. Thereby, a set of small measurements results often in an underestimation of the quantity of release, since these are positively related by $Q = \frac{\mu_m}{\phi^*}$. However, as discussed in Subsection 4.3.2, the decrease of dual concentration does not have a simple approximation, by which the effect of different measurement stations on the computation of quantity of release can not be quantified in advance of the source term reconstruction.

By this test case, relatively high measurements are noticed to have a great influence on the solution. As a result of diffusion, the retro plume spreads with a decreasing speed, resulting in high concentrated clouds being distributed in space after a period of time. In this way an high dual concentration is only possible in a relatively small area around the measurement station. Thereby, the reconstruction method expects an high dual concentration of the retro plume released from a station with a high measured quantity, since these values should



Fig. 5.2 Location probabilities for straight wind for reduction above 2.0 obtained by four different measurement grids. Each grid is represented by the measurement density given by the number of sampling stations per travel hour of the plume and longitude and latitude distance between sampling stations.

Grid	Dist. real	Quantity	Var.	Time interval	Max	Max
	loc. (km)	interval (Bq)	Qfactor		red.	prob. red.
1	258	[6.40E+14, 1.99E+15]	2.17	[4 July 17:00, 4 July 22:00]	9.7	0.34%
2	187	[9.36E+14, 1.09E+16]	3.07	[4 July 22:00, 5 July 2:00]	8.2	0.51%
3	51	[1.63E+15, 8.51E+15]	2.39	[5 July 10:00, 5 July 11:00]	14.1	2.10%
4	51	1.21E+16	3.01	5 July 11:00	12.6	7.07%
Source	0	1E+16		[5 July 9:00, 5 July 9:10]		

Table 5.3 Solutions with the highest RMSE reduction for straight wind case for four different measurement grids together with the actual source term (Source). Grid 1: latitude distance of 400km between stations, grid 2: 300km, grid 3: 200km and grid 4: 100km. This table contains distance to the actual source, time and quantity interval obtained by RSME reduction above 2.0, uncertainty in quantity of release and the maximum value of RMSE reduction and probability at this location. With uncertainty in distance of ± 33 km and in time of ± 1 hour.

correlate. A a result, a measurement station close to the source with a high measurement has a great influence on the solution. This can also have a negative influence when this small area is not covered by the PSL- grid because it was not chosen fine enough or when the weather conditions at that moment and time does not match the actual weather conditions. It is likely possible to correct for the influence of relatively high measurements by adding weights to the measurements and dual concentrations.

The solutions with highest RMSE reductions are shown in Table 5.3. In the second column the distance between the location with maximal RMSE reduction and the actual source location, Edinburgh in this case, can be found for all measurement grids. According to this table the method prefers locations close to the actual source and shows a better result for a finer measurement grid. The quantity and time intervals are obtained by the time period for which RMSE reduction exceeds the factor f = 2.0. Moreover, the table shows for each location another time interval, which is the result of the travel time of the retro plumes. At an early moment in time the proposed method is expected to compute higher release quantities than later in time, however the opposite is shown in the third column of Table 5.3. As was discussed earlier in this section, the low computed quantities are due the choice of used measurements and sampling locations. A set of low measurements provides an underestimation of the quantity of release. Although the quantities are low for the measurement grids 1,2 and 3, the computation of Q is quite stable for the best solutions for each measurement grid as can be seen by the variation of the Qfactor.

Since the source term is known: a 10 minutes release of 1×10^{16} Bq (Cesium-137) from Edinburgh at 5 July 9:00 AM, the reliability of the solutions can be discussed. In order to determine the accuracy of the solutions and how the distance and release quantity are

Layer	dt (hours)	$\ d\vec{x}\ _2$ (km)	Qfactor
1	≤ 6	≤ 108	\leq 4.47
2	≤ 12	\leq 216	≤ 5.09
:	•	:	÷
4	≤ 24	\leq 432	≤ 6.78
:	:	:	:
18	≤ 108	\leq 1944	\leq 35.22

Table 5.4 Layers definition in straight wind case

expected to progress over time, the used layers are defined and shown in Table 5.4. The expected Q factors are computed with measurements of the finest measurement grid 4 and are given by the following computation:

$$E_{Ofactor}(dt) = 5.16 \times 10^{-4} (2dt)^2 + 3.31 \times 10^{-2} |2dt| + 4.$$

The layers provides us with sorted solutions according to the performance, the frequency in those layers are shown by histograms in Figure 5.3. Grid 4 shows accurate results, most of the solutions are what is expected to find within 1 day difference from the real release moment¹. The underestimation of quantity of release highly influences the performance of the solutions, which is shown by the difference in dependency to quantity of grid 3 with respect to grid 4. Thereby, multiple possible solutions of grid 3 close to the actual source location, are descended in performance and by that fall into higher layers. Similarly, the performance of the solutions by grid 1 and 2 are also highly influenced by the underestimation of quantity of release. According to this can be concluded that a finer grid shows more accurate solutions, not only by distance and number of solutions, but also in quantity of release. However, the solutions of grid 4 may be more accurate if the PSL grid was chosen finer. The leading retro plume, the plume from the sampling location closest to the source, is expected to possibly travel between the locations of the possible source location grid. Because, in the first hours of release the cloud is really concentrated and may therefore not be noticed by the possible source location grid. From this can be concluded that there is a maximum on the achievement of accuracy obtained by increasing density of the measurement grid. Whenever this maximum is reached, the PSL grid should be chosen finer.

For more details about the frequencies shown in the histograms in Figure 5.3, in Table A.1 in the Appendix A the specific cumulative percentages of frequency per layer are presented for each measurement grid.

¹Note that the time differences are absolute values, so 1 day difference means in the period of 1 day before the release until 1 day after.

5.2 Synthetic cases



Fig. 5.3 Histograms with frequency of solutions in each layer and dependency to distance, quantity and time for each solution obtained by four measurement grids with a straight wind. Grid 1 is a measurement grid with around 400km distance in latitude between sampling stations, grid 2 with 300km, grid 3 with 200km and grid 4 with 100km distance.

5.2.2 Rotational wind

The previous case, which focuses on a wind blowing in one direction, is not realistic. On an average day the wind can be very unpredictable. Therefore, the following case focuses on a rotational wind field to determine how the method performs to chances in wind direction, incomplete weather information and measurements obtained multiple hours after the release started.

The wind field in this case produces a plume trajectory which shaped like a moving spiral, shown in Figure 5.4. Due to the rotation, a small area is covered by the plume and therefore the modelled period is shorter than for the straight wind field. Here an instantaneous release from Hanover in the period from 3 July 3:00 to 5 July 18:00 is simulated with a release of 1×10^{16} Bq Cesium-137 at the first time. Notice that sampling stations at the upper left from Hanover, would give reason to suspect that the release would be originated in the Netherlands



Fig. 5.4 Simulated release from Hanover integrated over time with a rotational wind and measurement locations (red stars) with a measurement density of 2.16.

or the United Kingdom, since the wind came in that direction. While stations in the South of the Netherlands would give reason to suspect in a similar way that the release was originated in the south. Accordingly, a naive solution based on wind directions at sampling stations are not sufficient to find an accurate source term in this case, although it would be sufficient in case of the straight wind field. Therefore, a dispersion model with weather simulation is essential for a varying wind field.

Even though, a measurement grid of 100 km provided us with an accurate source term reconstruction for the straight wind case, for this rotational case it was to rough. After the first test with an 100 km measurement grid was noticeable that with respect to the actual source term the quantity of release was underestimated, the time of release too early and by that the locations positioned later in the retro plume. This was likely the result of the use of to low and to few measurements, since high measurements highly influences the possible quantity of release. Because a measurement grid of 100 km grid was not sufficient fine, a measurement grid of 56 km is considered, with measurement density of 2.16.

For this test case three scenarios are discussed. The first scenario is when the actual wind field is known exactly at every hour, while in the second scenario the wind field is only known at every 3 hours. The third scenario is when measurements are only available after the first whole rotation of the plume of the simulated incident. That is, after the plume

Scenario	PSL's	Dist (km)	Time interval	Quantity interval (Bq)	Red. interval
1	48	366	[1 July 10:00, 3 July 02:00]	[2.86E+15, 2.53E+16]	[1.2, 2.7]
2	40	406	[1 July 18:00, 3 July 03:00]	[4.09E+15, 2.14E+16]	[1.2, 1.8]
3	603	753	[1 July 00:00, 4 July 15:00]	[6.68E+13, 3.09E+16]	[1.2, 4.6]

Table 5.5 General results of the method with factor 1.2 for a rotational wind in three scenarios. Scenario 1: full knowledge, scenario 2: partial weather knowledge and Scenario 3: a part of the measurements. This table contains the number of possible source locations, the maximal distance between two possible source locations and total time, quantity and reduction intervals. With uncertainty in distance of ± 65 km and in time of ± 1 hour.

crosses itself, such that the first five rows of the measurement locations from above shown in Figure 5.4, are not used in this scenario.

Unfortunately, in some scenarios no or very few possible source locations with an RMSE reduction exceeding 2.0 were obtained. For this reason, the solutions for this case are obtained by RMSE reductions exceeding 1.2. The possible source location areas are shown in Figure 5.5 together with the location probabilities. From this figure can be seen that less weather information does not show solutions that do not suffice, they are only slightly off the trajectory than was obtained from scenario 1. The main difference between these two scenarios is that all locations in scenario 2 are almost equally likely. By the RMSE reduction interval in Table 5.5 can thereby be noticed that the RMSE reductions are also much lower for scenario 2 than for the first scenario. Moreover, this case is on a local scale over a smaller period of time. On a larger scale with locally rotations of the wind incomplete weather information can have much more influence in the trajectory of the plume and even so on the solution field. As a result, a larger scale would apart from a change of plume trajectory, also show more uncertainty in the time of release and by that in the source location.

From the solution field of scenario 3, it is clear that less measurement stations show an higher uncertainty of the solution. However, since the wind field is known exactly, the locations with the highest probabilities are still in the trajectory of the actual plume and further from the actual source location the probability reduces quite fast. More details of the solution fields are shown in Table 5.5 with general results of the rotational wind case.

In Table 5.6 can be seen that the RMSE reduction is higher for scenario 3, even though the solutions of scenarios 1 and 2 are more probable. The reason for this is that scenario 3 has fewer measurement stations which are compared with the dual concentrations, which makes it easier to obtain a high RMSE reduction. Furthermore, this table shows that scenario 1 and 2 provide only one probable time and a high variation in the computation of quantity of release. This means that the RMSE reduction showed one narrow peak for those possible locations at the computed time. An example of these RMSE reductions over time are shown



Fig. 5.5 Location probabilities in case of a rotational wind with RMSE reduction factor 1.2 for all three scenarios. The scenarios are with (1) complete weather information, (2) weather information after every 3 hours and (3) only measurements after the first rotation of the simulated plume shown in Figure 5.4. Down right: Scenario 3 with only probabilities above 0.2%.

Scen-	Dist. Han-	Quantity	Var.	Time interval	Max	Max
ario	over (km)	interval (Bq)	Q		red.	prob. red.
1	31	9.2E+15	33.95	3 July 01:00	2.7	3.7%
2	31	6.21E+15	17.66	3 July 01:00	1.8	3.3%
3	27	[4.01E+14, 4.24E+15]	2.72	[2 July 17:00, 2 July 23:00]	4.6	0.41%
Source	0	1E+16		[3 July 3:00, 3 July 3:10]		

Table 5.6 Solutions with the highest RMSE reduction for rotational wind case in three scenarios together with the actual source term (Source). Scenario 1: full knowledge, scenario 2: partial weather knowledge and Scenario 3: a part of the measurements. This table contains the distance to the actual source, time and quantity interval obtained by RSME reduction above 1.2, uncertainty in quantity of release and the maximum value of RMSE reduction and probability at this location. With uncertainty in distance of ± 33 km and in time of ± 1 hour.

	Layer	dt (hours)	$\ d\vec{x}\ _2$ (km)	Qfactor
	1	≤ 6	≤ 108	≤ 5.09
	2	≤ 12	≤ 216	≤ 6.84
	:	:	•	:
	4	≤ 24	\leq 432	≤ 12.33
	:	•		÷
	14	≤ 84	≤ 1512	\leq 79.47
م	blo 57	I avora dafinit	ion in rotation	al wind an

Table 5.7 Layers definition in rotational wind case

in Figure A.3 in Appendix A. Thereby, a fast changing dual concentration at a possible source location over time, shows besides a RMSE reduction peak also a fast change in the computed amount of release. Due to discrete calculations of dual concentration in time and space, it is possible that other narrow peaks of RMSE reduction are not detected. This suggests that the PSL grid should have been chosen finer in order to detect better possible source terms. however, this scenario is actually not tested with a finer PSL grid. Furthermore, due to the rotation of the wind retro plumes pass by multiple locations twice, which results in multiple possible times of release. That is why scenario 3 in Figure A.3 shows for correlation and RMSE reduction multiple peaks in time at a location close to the actual source.

In order to show the performance of the solutions with respect to the actual source term, the layers shown in table 5.7 are used. The Qfactor is based on all measurements and given by

$$E_{Qfactor}(dt) = 2.30 \times 10^{-3} (2dt)^2 + 6.33 \times 10^{-2} |2dt| + 4.$$

The actual source term is in this case a release from Hanover at 3 July 00:00 of 10 minutes with a release of 1×10^{16} Bq Cesium-137.



Fig. 5.6 Histograms with frequency of solutions in each layer for three scenarios with a rotational wind. The dependency of the performance of the solutions are given by the percentage of solutions depending on respectively distance, difference in quantity of release and difference in time with respect to the actual source. The scenarios are with (1) complete weather information, (2) weather information after every 3 hours and (3) only measurements after the first rotation of the simulated plume shown in Figure 5.4.

Histograms of the frequency of solutions within the defined layers, are shown in Figure 5.6. Remarkable is that scenario 2 shows a higher percentage of solutions in each besides the first layer with respect to scenario 1, this can be seen more detailed in the frequency Table A.2 in Appendix A. This is likely because the trajectory in scenario 2 is only slightly off of the real trajectory. This results in a higher percentage of solutions in the lower layers, since scenario 2 has less possible source locations than scenario 1. Furthermore, scenario 3 shows many solutions in high layers, mostly due to time, however also due to high ratios between actual and computed release quantities. As can be seen in Table 5.6, scenario 3 with few measurement stations supports the conclusion that a set of low measurements underestimates the quantity of release.

5.2.3 Discussion of synthetic cases

From these synthetic cases we tried to examine how retro plumes and the model reacts on different sampling station positions and weather conditions.

First of all we have seen that a measurement density above 1 station per travel hour of the plume is enough, in case of a straight wind, to get an accurate solution field. In this case 95% of the solutions are within the source term boundaries of layer 4, which corresponds with a time difference of ± 24 hours from the actual source term. Furthermore, the solution of the finest measurement grid with highest RMSE reduction closely approach the actual source term. If less measurement stations are used, the solution area increase rapidly and the computed quantity of release decrease.

However, a straight wind is far from realistic. For this reason a local rotational wind is considered. From this case was noticeable that a finer measurement grid with around 2 stations per travel hour of the plume is required to get an accurate solution. Although the manually generated wind fields are really simple, with the same weather conditions for each location and a constant mixing layer, they still provided us with interesting results about computational errors due to discretization of time and space. The weather information from the dutch national weather service (KNMI) are provided for every 3 or 6 hours. From the experiment with weather information after every 3 hours was seen that the dispersion model has little problems with the real wind trajectory and by that, the method provided us a solution which was only slightly off from the real trajectory of the plume. However, this trajectory is most of all influenced by the discrete computation of dual concentrations. Since the quantity of the clouds are computed after every 10 minutes and discrete in space, local changes in wind direction and speed may not be observed, which can have large consequences after a long time period.

Furthermore, the dispersion model in general influences the outcome of the method. During the test cases no deposition was used in the dispersion model, since it required additional modelling to simulate a reversed rain in this Puff-model, which was out of scope of this study. Missing deposition is expected to detain the diffusion of radioactive materials, since everything stays within the simulated cloud. Decay also has a low influence during those experiments, because the used nuclide has a long decay half life. Although, for simulation of measurements and retro plumes the same nuclide (Cesium-137) is used, such that in both simulations the air pollutants decay slowly. The main components of the dispersion model are therefore wind speed and direction and diffusion of the cloud in horizontal and vertical directions.

However, the solution field also depends on the choice of the possible source location grid. Although, no experiments have been done with different PSL grids, the expectation is

that source locations close to the measurement locations require a fine PSL grid both in place and in time. This is because a retro plume close to the sampling location from which it was released, is small and concentrated. Such a small cloud of dual concentration, may not be noticed by a PSL grid with large space distances or large time steps.

In real life, among other wind conditions, a varying wind field is expected, thus a combination of the two manually generated wind fields. In order to find an accurate solution field, a measurement density of around 2 is expected to be enough. Notice that this highly depends on the weather conditions and how good the actual weather is approached in the dispersion model. The discretization of time and space results in computational errors, which are really small by a calm wind and by that results in a better source term reconstruction than by a varying wind field. Therefore, it is recommended to check the wind field in the time period of simulation beforehand to formulate expectations of the proposed reconstruction method. Accordingly, a suitable PSL grid can be chosen in such a way that many crucial wind changes are detected.

5.3 ETEX 1

Finally, the source term reconstruction method is tested on a real case, namely the first release from the European Tracer Experiment (also called ETEX)[27]. This tracer experiment was set up around 1990 by the European Commission (EC), the World Meteorological Organization (WMO) and the International Atomic Energy Agency (IAEA), which were later joined by the U.S. Department of Energy (USDOE). The purpose of this experiment was to test the response to an emergency, to organize the trace release and to collect data of measured air concentrations to investigate the performance of available dispersion models in order to improve these models.

Over the years many organizations in Europe joined this experiment, resulting in an sampling network of 168 ground-level sampling stations. Each sampling station was able to sample the air in periods over three hours, where each station started sampling delayed from West to East, while the release was in the West of France. For the choice of released materials it was most of all important to guarantee the safety for human and environment, stability in the air, low measurement background and high sensitivity of detection [27]. For that reason it was decided that perfluorocarbon compounds (PFCs) are being released in the air. To be specific, the actual material was perfluoromethylcyclohexane (PMCH) in case of the first experiment ETEX1.



Fig. 5.7 Sampling stations of ETEX. Black stations are used for the method since they has samples above the threshold 0.05 ng/m^3 . The measurement density is hereby 1.38 measurements per travel hour of the plume.

After four years of preparations the first tracer was released in October 1994. The actual source term was a release from 23 October 1994, 16:00 UTC until 24 October, 3:50 UTC. During this period a total of 340 kg PMCH were emitted.

First notice that the release was not instantaneous, for that reason the actual source term is expected not to be found exactly. Instead, we expect to find an averaged release per hour around the source location within the time interval of release. The quantity of release is per hour, because the measurements are averaged over three hours. Unfortunately, there was not enough time to adjust the reconstruction method for a continuous release, although it should be possible by small adjustments.

The measurements are sampled and averaged over 3 hours. For the test case only the first sampling intervals of measurement stations which measured quantity exceeding the threshold 0.05 ng/m³ are chosen to be used. This leaves us with the black colored sampling stations in Figure 5.7 and a measurement density of 1.38 measurements per travel hour of the plume. According to the synthetic cases, we expect that the number of measurements would be enough in case of an easy wind field. However, the measurement network is probably not fine enough to reconstruct the actual source location when there are sudden changes in wind directions.

After releasing retro plumes from all black measurement stations, the dual concentrations spreads as shown in Figure 5.8. From this figure can be seen that some sampling stations



Fig. 5.8 Dual concentration field of the used sampling stations (red stars) at five moments in time. From left to right: 25/10/1994 0:00, 24/10/1994 12:00, 23/10/1994 18:00, 23/10/1994 12:00, 23/10/1994 0:00.

provides retro plumes that differ highly from the yellow cloud centres. Those small diverging plumes results after a while in a large cloud of dual concentrations. However, the correlation computation of the method will highly reduce the possible source area, because the dual concentrations will be compared with the measurements.

Since the source term is known, the dispersion model can be used to simulate the measurements. In Figure 5.9 the real and modelled measurements are shown side by side, for each sampling location at the same sampling interval as was actually measured. From this figure can be seen that at some sampling locations the model does not expect measurements. The release of retro plumes from those stations anyway, likely leads to the diverging retro plumes shown in Figure 5.8. Thereby, the Root Mean Squared Error between those two measurements is $RMSE(\vec{\mu}_{real} - \vec{\mu}_{sim}) = 1.29$. More detailed figures of real and simulated measurements over time can be seen in Figure A.4 in Appendix A.



Fig. 5.9 Left: Real measurements at the first three hourly sampling interval for which concentration above threshold 0.05 ng/m^3 was measured. Right: Simulated measurements at the same moment as the real measurements. RMSE between those measurements is 1.29.

A computation of correlations between dual concentrations and the real and modelled measurements shows a bit different solution field. General information about those fields are shown in Table 5.8. The R in the first column of this table stands for the case with real measurements and S with simulated measurements. For the performance in this case a RMSE reduction factor of 1.2 is used, since the reductions do not exceed 2.0. From the table can be seen that the ideal case, with simulated measurements, is more specific about the solution. Since the solutions exists of less possible source locations, a shorter time period and smaller quantity interval. However, the maximum RMSE reduction of the simulated case is slightly smaller, which can be explained by the smaller range of measured quantities by the real measurements compared to the simulated measurements as can be seen in Figure 5.9.

With the use of real measurements, the reconstruction method provides the probability field with factor 1.2 as shown in figure 5.10. It seems that this solution exists of three possible source location fields with high RMSE reductions. One field close to the source in France, one below Ireland and the other one way further on the Ocean. However, notice that only a part of the measurements are used. As can be seen in Figure 5.7, sampling locations positioned west of the source location did not measure concentrations above 0.05 ng/m³. Since those sampling stations would have measured radioactivity if a source was positioned in the ocean, one could argue to only consider the possible source locations in France. Which would lead in this case to an accurate source term reconstruction.

Different from the real measurements, the simulated measurements provides us with a solution with a high preference for France. This is due to the stations that measured nothing



(a) Solution field obtained by real measurements. Three (b) Solution field obtained by simulated measurements.
 solutions with high RMSE reduction are highlighted.
 The solution with the highest RMSE reduction is highlighted.

Fig. 5.10 Location probabilities for ETEX1 for reduction above 1.2.

	PSL's	Dist (km)	Time interval	Quantity interval (ng)	Red. interval
R	534	1535	[23 Oct. 00:00, 24 Oct. 11:00]	[6.69E+12, 1.30E+18]	[1.2, 1.39]
S	171	1324	[23 Oct. 00:00, 24 Oct. 09:00]	[1.45E+13, 5.46E+14]	[1.2, 1.34]

Table 5.8 General results of the method with factor 1.2 for ETEX1 with real measurements (R) and simulated measurements (S). This table contains the number of possible source locations, the maximal distance between two possible source locations and total time, quantity and reduction intervals. The uncertainty in distance is given by ± 65 km and in time by ± 1 hour.

according to the model. The dual concentration of those stations do not have any influence on the correlation, since the dual concentration is multiplied with the zero measurement in the calculation of correlations. Thereby, the solution is only computed by the sampling stations that has measured during the simulated release. Retro plumes from these remaining sampling stations seems to split up into two when leaving France, which shows the gap in the solution field by simulated measurements in Figure 5.10. Furthermore, this difference between real and simulated measurements provides the additional solutions in case of the real measurements.

The solutions with highest RMSE reductions from this method are shown in Table 5.9. In this table, R1 is the best solution with real measurements, it is positioned in the area west of France in the Ocean. R2 represents the best solution with real measurement of the area just below Ireland and R3 the same for the area in France. Furthermore, S represents again the solution with the simulated measurements. To support the positioning of the solutions visually the locations are highlighted in Figure 5.1. Furthermore, Table 5.9 shows that the maximum RMSE reductions are higher in case of the real measurements, even though the simulated measurements are expected to show better solutions. A possible explanation is that the solutions R1, R2 and R3 highly correlates with measurements at the sampling stations which according to the dispersion model should not have measured radioactive materials in the air. Besides, the same table also shows that the locations close to the actual source have a high preference for the end of the actual release time interval. The likely reason for this is the increase of dual concentrations with time, since time is reversed in the dual dispersion model. An high dual concentration corresponds with an high possibility the source was positioned on that location in time according to the particular sampling station. Therefore a late moment in time with high dual concentration would be preferred over an earlier time with a lower dual concentration.

Since the method has a preference for the end of the release, the reconstructed solutions will be compared with the actual source term in the end of the actual release interval. The released quantity would then be divided over time which results in an averaged value per hour. Hereby the solutions will be compared with the actual source term existing of a release of 2.87×10^{13} ng (28.7 kg) at 24 October 1994 03:50 UTC from Monterfil.

Similar as with the two synthetic cases, layers are again used to order the solutions by performance with Qfactor defined by Equation 5.3 which gives the equation:

$$E_{Ofactor}(dt) = 4.26 \times 10^{-2} (2dt)^2 + 7.23 \times 10^{-2} |2dt| + 4.$$

This provides us with the layers shown in Table 5.10. The frequency of solutions in those layers can be seen in Figure 5.11, with the actual percentages shown in Table A.3 in Ap-

50	50						
	Dist. real	Quantity	Var.	Time interval	Max	Max	
	loc (km)	interval (ng)	Qfactor		red.	prob. red.	
R1	1142	[5.74E+14, 6.76E+14]	1.33	[23 Oct. 5:00, 23 Oct. 6:00]	1.39	0.206%	
R2	519	[9.77E+13, 2.97E+14]	0.77	[23 Oct. 17:00, 23 Oct. 19:00]	1.37	0.203%	
R3	58	[1.40E+13, 3.99E+13]	3.99	[24 Oct. 5:00, 24 Oct. 6:00]	1.35	0.201%	
S	120	1.42E+14	9.8E-7	24 Oct. 6:00	1.34	0.639%	
Source	0	2.87E+13		24 Oct. 3:50 (- 12 hours)			

Table 5.9 Solutions with the highest RMSE reduction for ETEX1 for real measurements(at locations R1, R2 and R3 as in Figure 5.10) and simulated measurements (S) together with the actual source term (Source). This table contains distance to the actual source, time and quantity interval obtained by RSME reduction above 1.2, uncertainty in quantity of release and the maximum value of RMSE reduction and probability at this location. Uncertainty in distance is given by ± 33 km and in time by ± 1 hour.

Layer	dt (hours)	$\ d\vec{x}\ _2$ (km)	Qfactor			
1	≤ 6	≤ 108	≤ 7.79			
2	≤ 12	≤ 216	≤ 20.45			
÷	:	:	:			
4	≤ 24	\leq 432	\leq 72.39			
:	:	:	•			
19	≤ 114	≤ 2052	\leq 1592.86			
Tab	Table 5.10 Layers definition for ETEX1					

pendix A. The solutions according to the real measurements are clearly more divided among the layers than in the ideal case with simulated measurements. Almost three percent of the solutions with real measurements falls in an higher layer due to errors in the computation of Q. These errors have the most effect for measurements close to the actual source. For example a probable amount of release of order 15 is computed at a location close to the actual source, instead of the actual release of order 13, such that the solution would now fall in Layer 5 instead of 1. Another 0.6% is bounded by the time, especially close to the actual source and all others by the distance with the actual source location. The histogram of the solutions by the real measurements then tells us that some solutions close to the source location. The simulated measurements provides more solutions close to the actual source location, although there are two diverging trajectories which cause many of the solutions being sorted in high layers. However, around 48% solutions still lies in the first four layers, which makes the solution almost accurate by our definition.



Fig. 5.11 Histogram of frequencies of solutions in each layer for ETEX1 with real measurements and simulated measurements. The dependency of the performance of the solutions are given by the percentage of solutions depending on respectively distance, difference in quantity of release and difference in time with respect to the actual source term.

	Dist. real	Quantity	Var.	Time interval	Max	Max
	loc (km)	interval (ng)	Qfactor		red.	prob. red.
R	11.4	1.51E+13	1.14	24 Oct. 6:00	1.21	0.179%
S	11.4	2.41E+14	2.73	24 Oct. 3:00	1.21	0.577~%
Source	0	2.87E+13		24 Oct. 3:50 (- 12 hours)		

Table 5.11 Actual source term (Source) and closest results to the actual source location for ETEX with real measurements R and simulated measurements S. This table contains distance to the actual source, time and quantity interval obtained by RSME reduction above 1.2, uncertainty in quantity of release and the maximum value of RMSE reduction and probability at this location.

In the end, the measurement positions and corresponding measurement density (1.38 measurements per travel hour of the plume) chosen for the case ETEX1 would have provided us with an almost accurate solution. Since simulated measurements provide us with a solution field with almost a majority of the solutions lying in layers 4 or lower. More sampling stations would likely show a more accurate solution field. However, the real measurements does not exactly match the modelled measurements, which results in more uncertainty of time and location. The difference between these measurements is among other things, likely due to exclusion of deposition from the dispersion model. Nonetheless, the simulated measurements represents the ideal measurements according to the dispersion model and by that provides us with the actual error between measurements and the model given by an RMSE of 1.29. As can be seen in Table 5.11, this difference in measurements between the real and modelled

world does not prevent the reconstruction method of presenting possible source terms closely to the actual source term.

Chapter 6

Conclusion and recommended research

6.1 Conclusion

In this thesis we proposed a source term reconstruction method to determine the most likely source term which clarifies the given measurements. For simplicity we assumed that the release was instantaneous. This assumption together with the definition of adjoint operators, provided us with an interesting relation between measurements and dual concentrations. This relation together with a method from Efthimiou and a solution scoring from IRSN resulted in the proposed reconstruction method. Therefore, a correlation-based reconstruction method is proposed which maximizes a Root Mean Squared Error reduction with respect to time and space. Furthermore, a probability field was used and the parameters to determine the performance of the method are defined.

With help of the synthetic cases it became clear that positioning of sampling stations highly influence the solution. An accurate solution field is defined to be a field with a majority of the solutions within expectations of a time interval of ± 24 hours with respect to the actual source term. The proposed method was according to the synthetic cases, expected to provide an accurate solution field for a sampling station grid with measurement density above 2.0 stations per travel hour of the plume. Few measurement stations provided a fast growing number of possible source locations. Furthermore, the values of the computed released quantities decreases with a decreasing number of stations, since high measurements are rare with few sampling stations. Thereby, was concluded that a set with low measurements provides solutions which underestimates the quantity of release.

However, instead of a measurement density of 2.0, ETEX1 used sampling stations and measurements with a measurement density of 1.38, which still provided an almost accurate solution field with simulated measurements. This performance is most likely due to the wide

range of simulated measurements. Nevertheless, the actual measurements had sampled lower quantities of radioactive material and as a result provided close to the actual source location lower computed quantity of release than by the simulated measurements.

As the KNMI provides wind fields every 3 or 6 hours, the incomplete weather information can influence the solutions of the proposed reconstruction method. Thereby, discretized calculations of dual concentrations results in computational errors. From the rotational wind case can be seen that partially known wind fields with a combination of discrete time and space steps can affect trajectories of retro plumes. The error of the trajectory is expected to amplify on a larger scale. For these reasons, the case ETEX1 shows a slightly diverged retro plume trajectory, which passed slightly north the actual source location. However, sampling stations which measured in real but not in the simulated case, provided multiple off trajectory retro plumes which results in additional possible source locations.

Partial since deposition is not included in the dispersion model and discretized calculations of measured concentration, the actual and simulated measurements show differences in sampled quantities. Multiple stations at the boundary which measured increments in real, do not have simulated measurements at that time and location. As a result of those measurements, the actual samples provide a solution field with a high uncertainty in source location and time of release. Although solutions in the neighbourhood of the actual source location approach closely to the actual source term at the end of the released time interval. Together with additional knowledge of zero measurements west of France near the coastline, could be argued to only use the possible source locations on land, which would result in a increased accurate source term reconstruction.

6.2 Recommended research

First of all, interesting further research would be to include deposition into the dual dispersion model. For example in case of rain, adding deposition into the simulation of retro plumes is expected to provide more accurate solutions by the reconstruction method.

Secondly, the proposed method can be generalized to an continuous release incident. As was discussed in Section 4.3, a continuous release adds a time integral to the calculation of RMSE reductions with respect to correlations. Computing the new RMSE reductions would require calculations of correlations and computed quantity of release continuously in time for multiple time lengths. The assumption of instantaneous release incidents provides solutions that prefers the end of the released time. For that reason, one could think of maximizing RMSE reduction with respect to release duration for releases ending at the time with the

highest correlation for each location. To test this idea, the hourly discretized correlations are approximately integrated over time. This first test provided an solution field with an overestimation of time of release, averaged expected quantity of release for each hour and highly scattered solutions in space. However, a more continuously calculation of correlations is expected to show less scattered solutions.

Furthermore, the cases are only discussed for a few different measurement grids and with one possible source location grid. Therefore, a more detailed research on those grids is recommended.

As with ETEX1, additional knowledge of sampling stations with zero measurements can provide restrictions to the possible source locations which increase the accuracy of the source term reconstruction. Thereby, additional research could be done on including zero measurements to the reconstruction method to restrict the possible sources. Other restrictions are possible by the chemical composition, since some compounds are characteristic for usage in particular kind of buildings such as hospitals or nuclear power plants.

Although high measurements can have a positive effect on the source term reconstruction, there are cases for which this is not true. As was seen from ETEX1, some sampling stations show high measurement errors which results in additional possible source locations. Therefore, it would be interesting to test the assignment of weights to measurements in such a way that the source term reconstruction method becomes less dependent on measurement and model errors.

Finally, the reconstruction method is not restricted to radioactive releases. This method is also suitable of every other air dispersion for which pollutants are measurable.

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Appendix A

Additional figures

A.1 Straight wind case

Grid 1: Density 0.11, 240 x 400 km



Grid 3: Density 0.38, 120 x 200 km

Grid 2: Density 0.19, 180 x 300 km



Grid 4: Density 1.48, 60 x 100 km



Fig. A.1 Measurement grids for straight wind case represented by measurement density and longitude and latitude distance between sampling stations.


Fig. A.2 Examples of correlation and RMSE reduction over time for two possible source locations with a straight wind.

Layer	grid 1	grid 2	grid 3	grid 4
	cum. %	cum. %	cum. %	cum. %
1	0.82 %	0.84 %	0.03 %	0.38
2	1.77 %	2.52 %	0.11 %	0.60
3	3.14 %	4.82 %	0.25 %	0.85
4	5.18 %	7.34 %	0.43 %	0.95
5	7.78 %	10.69 %	0.57 %	0.98
6	10.91 %	22.85 %	0.73 %	0.98
7	18.01 %	35.43 %	0.85 %	0.98
8	31.38 %	47.17 %	0.93 %	0.98
9	44.75 %	59.33 %	0.93 %	1.00
10	60.03 %	70.44 %	0.95 %	1.00
11	72.58 %	80.71 %	0.96 %	1.00
12	84.72 %	88.89 %	0.96 %	1.00
13	91.41 %	93.29 %	0.96 %	1.00
14	92.63 %	93.92 %	0.97 %	1.00
15	94.68 %	94.55 %	0.98 %	1.00
16	95.77 %	95.18 %	0.98 %	1.00
17	96.32 %	95.81 %	0.98 %	1.00
18	96.73 %	96.44 %	0.98 %	1.00
More	100.00 %	100.00 %	1.00 %	1.00

Table A.1 Cumulative frequency percentages in layers for straight wind for different grids. Respectively 400km, 300km, 200km and 100km distance in latitude.

A.2 Rotational wind

Layers	Scenario 1 %	Scenario 2 %	Scenario 3 %
1	18.75 %	25.00 %	2.82 %
2	41.67 %	55.00 %	8.46 %
3	70.83 %	82.50 %	15.59 %
4	77.08 %	90.00 %	21.06 %
5	85.42 %	100.00 %	26.37 %
6	95.83 %	100.00 %	41.46 %
7	100.00 %	100.00 %	59.20 %
8	100.00 %	100.00 %	87.06 %
9	100.00 %	100.00 %	90.71 %
10	100.00 %	100.00 %	93.86 %
11	100.00 %	100.00 %	94.86 %
12	100.00 %	100.00 %	96.19 %
13	100.00 %	100.00 %	97.18 %
14	100.00 %	100.00 %	97.35 %
More	100.00 %	100.00 %	100.00 %

Table A.2 Cumulative frequency percentages in layers with rotational wind for three scenarios. Scenario 1: complete information about the weather and all measurement stations, scenario 2: weather information only after every 3 hours and scenario 3: only measurements after the first rotation of the simulated plume.





A.3 ETEX1



(c) 24 October 18:00 - 21:00

(d) 25 October 18:00 - 21:00

Fig. A.4 Simulated concentration near the ground for actual source term of ETEX1 and real measurements (stars) sampled at four sampling intervals in time.

Layers	Real measurements	Simulated measurements
1	3.37%	12.28%
2	5.62%	29.24%
3	8.43%	42.11%
4	11.42%	47.95%
5	14.42%	56.14%
6	18.73%	66.08%
7	24.34%	77.19%
8	31.09%	84.80%
9	37.64%	91.23%
10	44.19%	95.32%
11	50.56%	100.00%
12	57.87%	100.00%
13	65.17%	100.00%
14	73.60%	100.00%
15	82.21%	100.00%
16	90.45%	100.00%
17	96.25%	100.00%
18	98.69%	100.00%
19	98.69%	100.00%
More	100.00%	100.00%

Table A.3 Cumulative frequency percentages in layers for ETEX1 with real and simulated measurements.