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**Adjoint backtracking  
for the verification of  
the CTBT**

J.-P. Issartel and  
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# Adjoint backtracking for the verification of the Comprehensive Test Ban Treaty

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## Abstract

An international monitoring system is being built as a verification tool for the Comprehensive Test Ban Treaty. Forty stations will measure on a worldwide daily basis the concentration of radioactive noble gases. The paper introduces, by handling preliminary real data, a new approach of backtracking for the identification of sources after positive measurements. When several measurements are available the ambiguity about possible sources is reduced significantly. As an interesting side result it is shown that diffusion in the passive tracer dispersion equation is necessarily a self-adjoint operator.

## 1. Introduction

We describe in this paper a new method for locating the source of an atmospheric tracer after concentration measurements in the air. The method is based upon an interpretation of the adjoint transport equation as an inverse transport equation. It applies to passive tracers or to tracers subject to some linear decay processes such as radioactive decay or rain scavenging. The connection of backtracking with the adjoint transport equation has been mentioned long ago by several authors (Pudykiewicz, 1998; Uliasz and Pielke, 1991). In a first paragraph we endeavoured nevertheless, in order to better handle this connection, to establish it on theoretical bases. Each measurement is taken into account through its retroplume representing the air of the sample scattered back in time according to a dispersion equation which is both adjoint or inverse. The retroplumes associated to several measurements may be combined together in order to reveal in a non statistical quantitative manner which sources are compatible with the measurements.

Our questions originally arose in the context of the Comprehensive Test Ban Treaty (Hourdin and Issartel, 2000). An international system, made up of forty stations, will provide a worldwide monitoring of radioactive gases produced by the tests.  $^{133}\text{Xe}$ , the

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main one, may be released as well by nuclear plants (Kunz, 1989) so that ambient concentrations may interfere with the detection of nuclear tests. After underwater tests  $^{133}\text{Xe}$  reaches quickly the atmosphere. It exhales through faults during tenths of hours after underground tests. An atmospheric test would jointly release radioactive aerosols.

Such aerosols are accurately monitored, in the frame of the Treaty, by an eighty station network. Hence, when  $^{133}\text{Xe}$  is detected in the absence of aerosols the source to be identified is probably a fixed point at ground or sea level from where the gas possibly spread during hours. Only such sources will be considered hereafter. They are met in a lot of industrial circumstances (Wotawa et al., 1998).

The method will be presented with data produced by the CTBT station of Freiburg im Breisgau, Germany, where several types of detectors were calibrated during 2000. A  $100 \text{ mBq}\cdot\text{m}^{-3}$  peak of  $^{133}\text{Xe}$ , ten times above the background, was detected on 3 February 2000, in a sample taken by a U.S. detector between 02:00 and 10:00 UT. It has not been possible to confirm any source for this event.

The calculations were performed with the atmospheric transport model POLAIR (Sportisse et al., 2002; Sartelet et al., 2002) developed at the Centre d'Enseignement et de Recherche Eau, Ville, Environnement. POLAIR is the fruit of a close cooperation with the team in charge at Electricité de France of the passive atmospheric transport model Diffeul (Wendum, 1998). It is a fully modular three dimensional Eulerian chemistry transport model. Advection is solved with a flux limiter method; diffusion is solved by a three point scheme. The reactive part of the model was switched off for the present application. In order to cover western Europe we used a grid extending from  $15^\circ$  W to  $35^\circ$  E and from  $35^\circ$  N to  $70^\circ$  N with outer clean air boundary conditions. The horizontal resolution of the model was  $0.5^\circ \times 0.5^\circ$  with fourteen Cartesian levels at 32, 150, 360, ..., 6000 m above ground or sea level and a 15-min time step. Meteorological data produced by the European Centre for Medium Range Weather Forecast were kindly supplied by Météo France. These six-hourly data had the same horizontal resolution as POLAIR but had to be interpolated according to the vertical Cartesian levels of the model.

## 2. Inverse transport

We consider in this section the release, dispersion and measurement of a tracer passively transported by the motion of the air. All information about this passive transport between parts of the atmosphere may be summed up by an exchange rate. Let's consider two volumes  $S$  at time  $t_s$ ,  $\mathcal{D}$  at time  $t_d$ ,  $t_s \leq t_d$ . In practice these volumes will be in our model the grid meshes of the source of tracer and of the detector. The mass of the air contained in  $S$  at  $t_s$  and  $\mathcal{D}$  at  $t_d$  are denoted  $M_s$ ,  $M_d$ , the mass of all air particles exchanged by the two volumes in the prescribed delay is denoted  $M_{ex}$ . The exchange rate is defined as:

$$\varepsilon(S, t_s, \mathcal{D}, t_d) = \frac{M_{ex}}{M_s M_d} \quad (1)$$

This rate equally describes the dispersion of the air from  $S$  or the origin of the air in  $\mathcal{D}$ . Normal, forward transport and this backtracking are somehow equivalent and accordingly the analytic description of the second will be readily deduced from that of the first. An amount  $q$  of tracer released in  $S$  at  $t_s$  generates a plume with an average concentration per unit mass of air  $q \varepsilon(S, t_s, \mathcal{D}, t_d)$  measured in  $\mathcal{D}$  at  $t_d$ . The same amount  $q$  released in  $\mathcal{D}$  at  $t_d$  and transported back in time generates a 'retroplume' with the same average concentration per unit mass  $q \varepsilon$  in  $S$  at  $t_s$ .

Let denote  $c(\mathbf{x}, t)$  the local concentration per unit mass of air after  $t_s$  of the plume of air from  $S$  and symmetrically  $c^*(\mathbf{x}, t)$  the concentration before  $t_d$  of the retroplume of the air sampled in  $\mathcal{D}$ . We obtain (Hourdin and Issartel, 2000) a reciprocity relation:

$$\bar{c}(\mathcal{D}, t_d) = \bar{c}^*(S, t_s) \quad (2)$$

where the overbars stand for volume average. For  $q = 1$  direct and inverse concentrations per unit mass of air are subject to:

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c + \zeta(c) = \sigma \quad (3)$$

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$$-\frac{\partial c^*}{\partial t} - \mathbf{v} \cdot \nabla c^* + \zeta(c^*) = \pi \quad (4)$$

$\mathbf{v}$  is the wind field.

The release and the sampling of amounts of air  $M_s$  in  $\mathcal{S}$  at  $t_s$ ,  $M_d$  in  $\mathcal{D}$  at  $t_d$ , both normalised according to the requirement  $q = 1$ , are accounted for by the following source and detector functions; the symbol  $\delta_{\text{time}}$  is a time Dirac so that its physical unit is the inverse unit of time:

$$\sigma(\mathbf{x}, t) = \frac{\delta_{\text{time}}(t - t_s)}{M_s} \quad \text{in } \mathcal{S}, \quad 0 \text{ outside} \quad (5)$$

$$\pi(\mathbf{x}, t) = \frac{\delta_{\text{time}}(t - t_d)}{M_d} \quad \text{in } \mathcal{D}, \quad 0 \text{ outside} \quad (6)$$

In a more general situation a source function may be largely spread in space and time, for instance the source of carbon dioxide. This is true as well for the detector function. The detector may deliver time averaged measurements corresponding to the time interval when the sample was taken, it may furthermore be airborne with a varying position. As for the units, they are the same, here, for  $\sigma(\mathbf{x}, t)$  and  $\pi(\mathbf{x}, t)$  both obtained as rates per unit mass of ambient air and per unit time with the unit amount of air released or sampled switched off by the normalising requirement  $q = 1$ .

The diffusion operator  $\zeta$ , unlike the winds, has the same sign in Eqs. (3) and (4): diffusion symmetrically dilutes the fate of  $\mathcal{S}$  and the origin of  $\mathcal{D}$ . The reason is the statistical time symmetry of microscopic turbulent motions averaged into a macroscopic diffusion. The obstacle of an unphysical anti-diffusion, classically restricting backtracking to the Lagrangian investigation of individual backtrajectories, is avoided in this Eulerian approach.

Equation (4) rebuilds a macroscopic history of the air sampled by the detector. It is as well a sensitivity equation, i.e. an adjoint equation as we now explain. Notice that

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the measurement behaves as a scalar product of the tracer concentration and of the detector function:

$$\mu(c, \pi) = \int_{\Omega \times T} \rho(\mathbf{x}, t) c(\mathbf{x}, t) \pi(\mathbf{x}, t) d\mathbf{x} dt \quad (7)$$

The integration is over the atmosphere  $\Omega$  and the time domain  $T$ .  $\rho$  is the density of the air. Let  $\mathcal{L}$  and  $\mathcal{L}^*$  be the linear operators defined by the forward and backward Eqs. (3) and (4) (or Eqs. 10 and 11) together with adequate zero boundary conditions:  $c = \mathcal{L}(\sigma)$ ,  $c^* = \mathcal{L}^*(\pi)$ . The measurement  $\mu$  tied to any source  $\sigma$  and sampling distribution  $\pi$  decomposes according either to elementary samples,  $\delta\mu_x = \rho c \pi(\mathbf{x}, t) d\mathbf{x} dt$ ,  $\mu = \int \delta\mu_x$ , or to elementary releases  $\delta\mu_y = \rho \sigma c^*(\mathbf{y}, u) d\mathbf{y} du$ ,  $\mu = \int \delta\mu_y$ . Hence we obtain a general form of the reciprocity relation (2):

$$\int_{\Omega \times T} \rho \mathcal{L}(\sigma) \pi(\mathbf{x}, t) d\mathbf{x} dt = \int_{\Omega \times T} \rho \sigma \mathcal{L}^*(\pi)(\mathbf{y}, u) d\mathbf{y} du \quad (8)$$

The relation shows how source and detector change roles. As announced the operators  $\mathcal{L}$  and  $\mathcal{L}^*$  are adjoint for the measurement product and so are Eqs. (3) and (4).

The analytic form of the measurement product is mainly conventional. It would become  $\mu = \int_{\Omega \times T} C \pi d\mathbf{x} dt$  with a concentration of tracer  $C = \rho c$  referred to the unit volume of air. But then, with  $\pi$  a sampling rate still referred to the unit mass of ambient air, so would be the adjoint concentration  $c^*$  unlike  $C$ . The symmetry of normal and adjoint transport would be hidden and so would be the interpretation of the latter as an inverse transport.

In reactor and neutron transport theory  $c^*(\mathbf{x}, t)$  is called the ‘importance’ for the measurement of a particle released in  $\mathbf{x}$  at  $t$  (Lewins, 1965).

The adjoint interpretation of inverse transport implies that, for appropriate conventions, diffusion is self-adjoint:

$$\zeta = \zeta^* \quad \text{i.e.} \quad \int \rho c \zeta(c^*) d\mathbf{x} dt = \int \rho \zeta(c) c^* d\mathbf{y} du \quad (9)$$

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This property may be compared to the similar property of the linearised diffusive collision operator of the Boltzmann transport equation for particles in the position-velocity space of kinetic theory (McCourt et al., 1990).

The self-adjoint constraint is fulfilled by the classical Fickian gradient diffusion used in POLAIR with a coefficient  $\kappa$ :  $\zeta(c) = -\frac{1}{\rho} \nabla \rho \kappa \nabla c$ .

During its transport by the motions of the air  $^{133}\text{Xe}$  undergoes a linear decay. Its half life  $\tau_{1/2} = 5.5$  days corresponds to a constant  $\lambda = \log 2 / \tau_{1/2}$ . It is possible to take this decay into account for the backward calculations. Just like diffusion it has exactly the same effect in the inverse world as in the direct world:  $c^*$  decays towards the past the same way as  $c$  decays towards the future. This may be surprising but merely means that, because of the losses of tracer, the importance of ancient sources for the measurement is attenuated. Forward and backward transport equations associated to  $^{133}\text{Xe}$  with a vertical gradient diffusion read as:

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c - \frac{1}{\rho} \frac{\partial}{\partial z} \left( \rho \kappa \frac{\partial c}{\partial z} \right) + \lambda c = \sigma(\mathbf{x}, t) \quad (10)$$

$$-\frac{\partial c^*}{\partial t} - \mathbf{v} \cdot \nabla c^* - \frac{1}{\rho} \frac{\partial}{\partial z} \left( \rho \kappa \frac{\partial c^*}{\partial z} \right) + \lambda c^* = \pi(\mathbf{x}, t) \quad (11)$$

### 3. A single measurement

When on 3 February 2000, a peak of  $100 \text{ mBq} \cdot \text{m}^{-3}$  was detected, the question of its origin immediately arose. Such a question is generally answered in terms of Lagrangian backtrajectories: the wind field  $\mathbf{v}(\mathbf{x}, t)$  is integrated backward departing from the detector at a time related to the detection. As a result a curve is obtained supposedly passing by the real source.

In order to account for the duration of the measurement, eight hours in Freiburg, or for the random effect of diffusion, the previous calculation would be repeated many times. This amounts to calculating back in time the trajectories of many Lagrangian particles.

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It is often considered that, if many backtrajectories go back to a certain region, then the source is probably there.

In fact calculating many backtrajectories amounts to calculating the concentration  $c^*(\mathbf{x}, t)$  of the retroplume emitted back in time by the detector. If we investigate the origin of a single particle sampled by the detector, the probability density (per unit mass of air) of its past position is indeed  $c^*$  calculated for a normalised detector function (the total amount of adjoint tracer released is normalised to the unity). Nevertheless when macroscopic sources are investigated this statistical interpretation of the retroplume is erroneous. If many backtrajectories go back to a certain region this just means that the region contributed much air to the sample and a source there should not be very big in order to account for the measurement.

A source of intensity  $Q$  in  $\mathbf{x}$  at  $t$  will generate a measurement  $\mu = c^*(\mathbf{x}, t) Q$ . In other words a measurement  $\mu$  can be explained by an instantaneous point source in  $\mathbf{x}$  at  $t$  of intensity  $Q = \frac{\mu}{c^*(\mathbf{x}, t)}$ . The retroplume establishes a constraint between the position of the source and its intensity. This analysis easily extends to the case of point sources that are not instantaneous. Suppose a source in  $\mathbf{x}$  has a rate of release  $D(t) \geq 0$  per unit time. The total release is  $Q = \int D(t)dt$  and the measurement is  $\mu = \int c^*(\mathbf{x}, t)D(t)dt$ . As  $D$  is a non-negative function  $\mu \leq \max_t c^*(\mathbf{x}, t) \int D(t)dt$ , or:

$$Q \geq \frac{\mu}{\max_t c^*(\mathbf{x}, t)} = Q_{\min}(\mathbf{x}) \quad (12)$$

We still do not know where the source actually lies. It could lie in any position  $\mathbf{x}$  provided the retroplume went there at some moment. Nevertheless the threshold function  $Q_{\min}$  shows that not all positions are equivalent. A source far away from the detector should be greater than a close one.

The retroplume of the peak measurement of  $^{133}\text{Xe}$  in Freiburg has been calculated by model POLAIR according to Eq. (11) with a normalised detector function  $\pi$  concentrated in Freiburg at position  $\xi_F$ , and during an eight hour interval  $\Delta t$ ; the symbol  $\delta_{sp}$  is

a space Dirac so that its physical unit is the inverse unit of volume:

$$\pi(\mathbf{x}, t) = \begin{cases} \frac{\delta_{sp}(\mathbf{x}-\xi_F)}{\Delta t \rho(\xi_F, t)} & 2 \leq t \leq 10 \text{ UT, } 2000/02/03 \\ 0 & \text{otherwise} \end{cases} \quad (13)$$

We have also calculated a traditional Lagrangian backtrajectory upstream from the detector. As POLAIR is fundamentally a Eulerian model, we performed this Lagrangian calculation by just setting horizontal and vertical diffusions to zero. Considering Fig. 1 we see that on 2 February 2000, the retroplume and this backtrajectory move to the northwest. During the morning of that day the retroplume turns to the southwest mainly above the Atlantic. The Lagrangian backtrajectory moves to the southwest too, but towards Gascony far away from the average position of the retroplume. Moreover, as can be seen on Fig. 2a, an industrial source in Gascony should be as big as  $Q_{\min} = 1000 \text{ TBq}$  which is orders of magnitude above the possible exceptional releases by nuclear civilian installations. It seems more reasonable to investigate industrial sources northwest of Freiburg. So far, no such source has been confirmed. The hypothesis has been proposed that the  $100 \text{ mBq}\cdot\text{m}^{-3}$  peak was related to a very weak medical source in Freiburg. Indeed German hospitals use  $^{133}\text{Xe}$  for pulmonary investigations. A nuclear test generates  $1000 \text{ TBq}$  of  $^{133}\text{Xe}$  per kiloton, only 10% exhale in the case of an underground explosion. Regardless of other considerations the  $Q_{\min}$  calculated for most western Europe would be compatible with a 10 kiloton test.

#### 4. Several measurements by a single station

The diagnosis above can be improved so as to determine minimum total releases  $K_{\min}(\mathbf{x})$  taking into account the constraints imposed by several measurements obtained in Freiburg. The constraining nature of these additional observations is easy to understand: if the source lies far away, the plume of  $^{133}\text{Xe}$  will have much broadened before

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reaching Freiburg and it will be detected during a long time. If successive measurements in Freiburg display a narrow peak, the source cannot be too far away. In order to evaluate this effect we handled the following four observations with the peak measurement now being labelled 1:

- 5  $\mu_0 = 6 \text{ mBq.m}^{-3}$  from 18 to 2 UT, 2000/02/02-03  
 $\mu_1 = 103 \text{ mBq.m}^{-3}$  from 2 to 10 UT, 2000/02/03  
 $\mu_2 = 42 \text{ mBq.m}^{-3}$  from 10 to 18 UT, 2000/02/03  
 $\mu_3 = 6 \text{ mBq.m}^{-3}$  from 18 to 2 UT, 2000/02/03-04

10 To each measurement a retroplume  $c_i^*(\mathbf{x}, t)$  may be related. A source in  $\mathbf{x}$  with a rate of release  $D(t) \geq 0$  now undergoes the four constraints:

$$\mu_i = \int c_i^*(\mathbf{x}, t) D(t) dt \quad i = 0, 1, 2, 3 \quad (14)$$

We considered in fact the following system of constraints were possible errors are widely taken into account:

$$D(t) \geq 0$$

- 15  $0 \text{ mBq.m}^{-3} \leq \int c_0^*(\mathbf{x}, t) D(t) dt \leq 10 \text{ mBq.m}^{-3}$   
 $52 \text{ mBq.m}^{-3} \leq \int c_1^*(\mathbf{x}, t) D(t) dt \leq 206 \text{ mBq.m}^{-3}$   
 $21 \text{ mBq.m}^{-3} \leq \int c_2^*(\mathbf{x}, t) D(t) dt \leq 84 \text{ mBq.m}^{-3}$   
 $0 \text{ mBq.m}^{-3} \leq \int c_3^*(\mathbf{x}, t) D(t) dt \leq 10 \text{ mBq.m}^{-3}$

20 We want to determine the minimum value  $K_{\min}(\mathbf{x})$  of the total release  $\int D(t) dt$  among all admissible rate functions  $D(t)$ . This linear optimisation problem can be easily managed, when time is discretised, by means of the so called ‘simplex’ algorithm. This classical algorithm was first described by Dantzig (1963). We operated it locally, for each position  $\mathbf{x}$  at ground or sea level in western Europe and for sources starting from

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25 January. The results are reported on Fig. 2b. For most positions the above constraints are not compatible. This means that the  $^{133}\text{Xe}$  detected in Freiburg cannot have originated there. The threshold function  $K_{\min}(\mathbf{x})$  is clearly more restrictive than  $Q_{\min}$  and the new diagnosis clarifies the previous one. Admissible sources now lie in a narrow strip departing from Freiburg to the northwest through France, Belgium, Great Britain and terminating one thousand kilometres off Ireland. Industrial sources should not be sought further than Wales. The diagnosis clearly excludes the southern part of France were the previous one already allowed only prohibitively big sources. A real advantage is obtained in the western part of France and southern part of England where sources as large as some tenths of kilotons, previously admissible, are now excluded.

Notice that a weak source close to Freiburg is not excluded. The reason is that when all measurements are from a single station, the same number of local contaminations make up an admissible source. Therefore, a single station will never be in a position to exclude a source in its close environment. This difficulty can be partly removed if we assume a limited duration of the release. The duration of industrial releases is classically less than twelve hours, one working day. Such a signal, emitted in the neighbourhood of Freiburg, should not interest more than three eight hour samples. It is nevertheless more convenient to use the local optimisation method with information from several stations.

## 5. Measurements from several stations

The local optimisation method described above is just an abridged way to handle the information contained in the retroplumes. A more accurate understanding of the meteorological situation may require a complete analysis as we now explain. The event detected on 3 February 2000 was observed only in Freiburg. The CTBT station of Stockholm was not yet operating. Let's just imagine what could have been said if a 24 hour sample had been taken there on 3 February 2000. We denote the corresponding retroplume as  $c_S^*$  and the fictitious measurement as  $\mu_S$ . A source in  $\mathbf{x}$  with a rate of

release  $D(t)$  would thus lead to  $\mu_S = \int c_S^*(x, t) D(t) dt$ .

We imagined two situations. Firstly the requirement that  $\mu_S = 0 \text{ mBq.m}^{-3}$  has been combined with real data from Freiburg in the local optimisation. This additional information would not change the final result. This means that sources previously diagnosed would be mostly compatible with a zero valued measurement in Stockholm in such a way that Fig. 2b is unaltered. Secondly the requirement that  $\mu_S = 100 \text{ mBq.m}^{-3}$  would clearly exclude weak sources close to Freiburg as shown by Fig. 2c. Only big sources northwest of England would now be acceptable.

We now place the transparent figure representing the retroplume  $c_S^*$  from Stockholm on top of the figure representing the peak retroplume  $c_1^*$  from Freiburg. Considering the resulting Fig. 3 we appreciate the connections of each point in space and time with the two measurements. We first notice that the retroplumes intersect marginally. This is the reason why a zero valued measurement in Stockholm does not alter the local optimisation diagnosis. We still notice that the retroplume from Stockholm does not meet western continental Europe. A source there could not contaminate the sample of Stockholm and would be excluded by the local optimisation diagnosis for a virtual measurement  $\mu_S = 100 \text{ mBq.m}^{-3}$ . In that case we notice furthermore that instantaneous spot sources may be considered only in Scotland where the retroplumes intersect marginally. Other acceptable sources should have a duration greater than twelve hours corresponding to the delay separating the passings of one and other retroplumes over most positions.

## 6. Conclusions

Among the forty CTBT noble gas stations many are settled in industrial areas close to civilian sources of  $^{133}\text{Xe}$  such as nuclear plants or hospitals. So, on the one hand, it is to be expected that sometimes several stations close to each other will simultaneously detect abnormal concentrations corresponding to independent local events. On the other hand, nuclear tests would certainly be seen by many stations but they are highly

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unlikely. The above method enables to discriminate between these two circumstances. This is a real asset with respect to the diplomatic and political aims of the CTBT. Indeed a set of positive data produced by independent local events will hardly be compatible with any single point source. The local optimisation method will explore this compatibility in any set of data and point it out as a worrying criterion while restricting the possible compatible area to worry about.

The methods proposed in this paper pertain to a now flourishing domain of inverse problems with many new ideas (Ternisien et al., 2000). Many studies are currently published about a number of atmospheric species (van Aardenne et al., 2001; Sportisse and Quélo, 2002). These methods generally aim at rebuilding a complex source  $\sigma(\mathbf{x}, t)$  by means of concentration measurements. The information contained in such measurements  $\mu_k$  may be summarised into the following equations by means of the associated adjoint retroplumes  $c_k^*$ :

$$\mu_k = \int \rho(\mathbf{x}, t) c_k^*(\mathbf{x}, t) \sigma(\mathbf{x}, t) d\mathbf{x} dt \quad (15)$$

This equation or system of equations is linear with respect to the source  $\sigma$  and its inversion as such has been proposed by Seibert (2001) in the frame of the CTBT. Such inversions may be considered data assimilation techniques because the system 15 is generally drastically under-determined. Considering again that the measurement process defines a scalar product we see that measuring the  $\mu_k$  amounts to determine the orthogonal projection of the function  $\sigma(\mathbf{x}, t)$  over the retroplumes  $c_k^*(\mathbf{x}, t)$ . The real source  $\sigma$  cannot be determined exactly but the available information enables to propose some linear combination of the  $c_k^*$  as an estimation for it. A general theory of such inverse problems, especially the regularisation of the estimation by a ‘truncated singular value decomposition’ (TSVD), has been addressed by Bertero et al. (1985, 1988) in a context dominated by image deblurring purposes.

This is not the approach that has been followed in this paper. We did not try to determine a source  $\sigma$ , we endeavoured to determine the position  $\mathbf{x}$  of a point source. The system 15 is linear with respect to  $\sigma(\mathbf{x}, t)$ , not with respect to  $\mathbf{x}$ . In order to explore

which positions  $\mathbf{x}$  were possible positions for a point source, we have degraded the complete linear system 15 into local systems:

$$\mu_k = \int c_k^*(\mathbf{x}, t) D(t) dt \quad (16)$$

We used then the linearity of the local systems with respect to a positive local source 5  $D(t)$  to build criteria  $Q_{\min}(\mathbf{x})$  or  $K_{\min}(\mathbf{x})$  that are clearly non-linear functions of the position.

In the frame of the treaty the calculation of a complex source by linear assimilation techniques would be of interest in order to confirm that a set of positive measurements is due to several local events. An important challenge for this assimilation should be to 10 take into account the non-negative constraint:  $\sigma(\mathbf{x}, t) \geq 0$ . This theoretic aim has been investigated by de Villiers et al. (1999).

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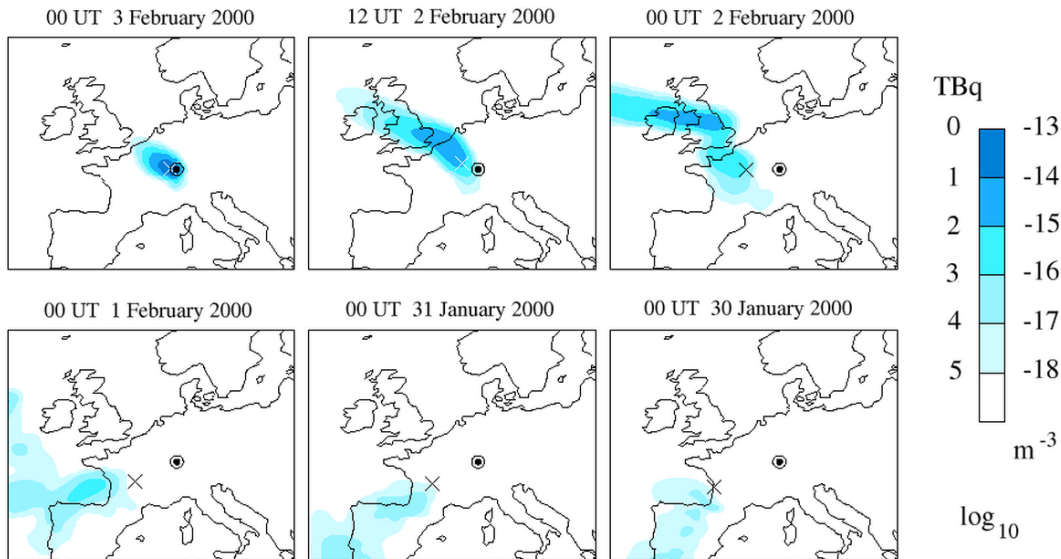
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**Fig. 1.** Retroplume  $c_1^*(\mathbf{x}, t)$  at ground or sea level corresponding to the  $100 \text{ mBq.m}^{-3}$  peak in Freiburg between 02:00 and 10:00 UT on 3 February 2000. In theory  $c_1^*$  should be given per kg of air but at ground level it is roughly equivalent and more convenient to give it per  $\text{m}^3$ . The same figure may be read in terms of possible point sources  $q = 100 \text{ mBq.m}^{-3} / c_1^*(\mathbf{x}, t)$  with a scale in TBq of  $^{133}\text{Xe}$ . The circle on the images indicates the position of Freiburg. The cross describes the backtrajectory of a Lagrangian particle departing back in time from Freiburg on 3 February 2000 at 06:00 UT. Notice that the Lagrangian particle does not follow the center of the Eulerian retroplume.

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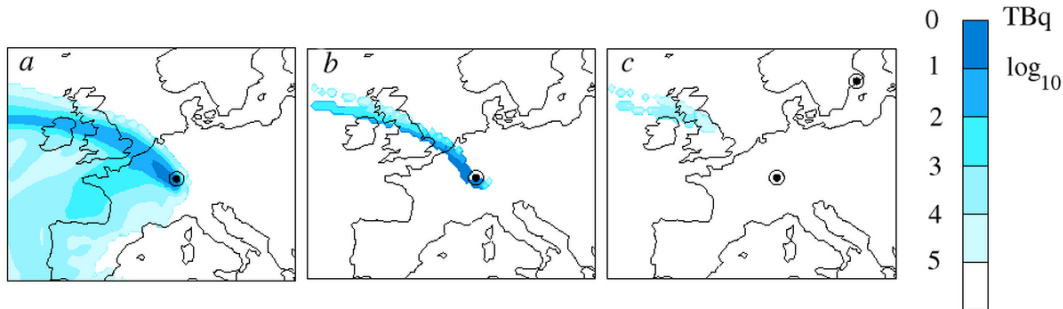
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**Fig. 2.** Minimum release from a point source at ground or sea level **(a)**  $Q_{\min}(\mathbf{x})$  compatible with the peak measurement of Freiburg  $\mu_1 = 103 \text{ mBq.m}^{-3}$  **(b)**  $K_{\min}(\mathbf{x})$  compatible with the series of measurements in Freiburg  $\mu_0 = 6$ ,  $\mu_1 = 103$ ,  $\mu_2 = 42$ ,  $\mu_3 = 6 \text{ mBq.m}^{-3}$ ; this value of  $K_{\min}(\mathbf{x})$  would not be altered by considering a fictitious measurement in Stockholm  $\mu_S = 0 \text{ mBq.m}^{-3}$  on 3 February 2000 **(c)**  $K_{\min}(\mathbf{x})$  compatible with the series of Freiburg completed with a fictitious measurement  $\mu_S = 100 \text{ mBq.m}^{-3}$  in Stockholm.

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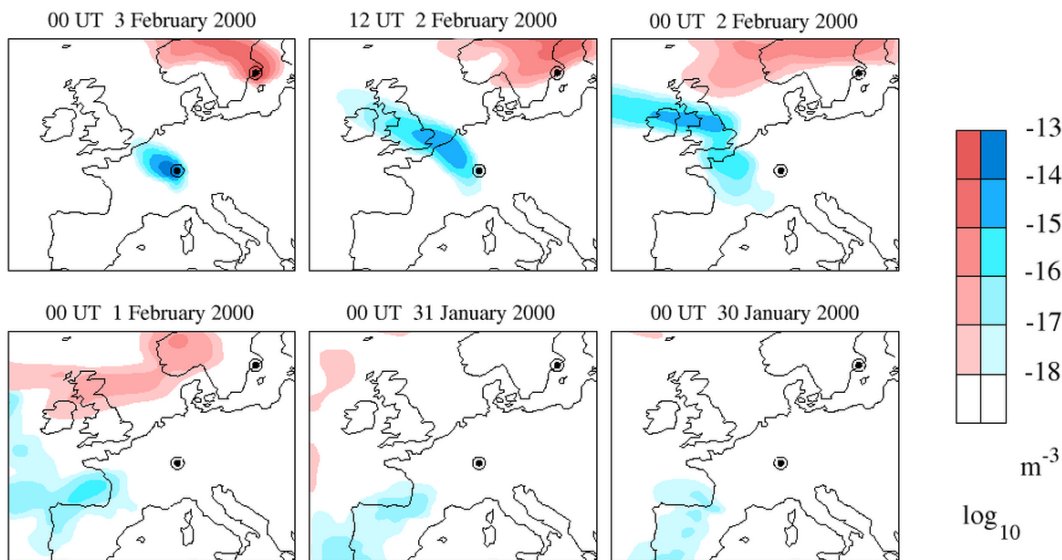
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**Fig. 3.** In blue the retroplume  $c_1^*$  associated to the peak detection of  $^{133}\text{Xe}$  in Freiburg. In red the retroplume emitted by the virtual measurement sampled in Stockholm between 00:00 and 24:00 UT on 3 February 2000. The retroplumes intersect only marginally in Scotland. The measurements are fundamentally independent. Scotland is in fact the only possible position for an instantaneous point source to contaminate positively both measurements. Both retroplumes flow over Ireland or above the ocean West of Ireland but never simultaneously. A point source there could contaminate both measurements but it could not be an instantaneous source.

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