A minimum curvature algorithm for tomographic reconstruction of atmospheric chemicals based on optical remote sensing

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Abstract. Optical remote sensing (ORS) combined with the computerized tomography (CT) technique is a powerful tool to retrieve a two-dimensional concentration map over the area under investigation. But unlike the medical CT usually uses, the beam number used in ORS-CT is usually dozens comparing to up to hundreds of thousands in the former. ORS-CT usually uses a beam number of dozens, which thus severely limiting the spatial resolution and the quality of the reconstructed map. This situation makes the smoothness' a priori information is therefore crucial especially necessary for ORS-CT. Algorithms produce smooth reconstructions include smooth basis function minimization (SBFM), grid translation and multiple grid (GT-MG), and low third derivative (LTD), among which the LTD algorithm is a promising one withbecause of fast speed and simple realization. But However, its characteristics and the theoretically basis are not clear must be clarified to better understand the characteristics of its smoothness constraints. Moreover, the computational efficiency and the reconstruction quality need to be improved for practical applications. This paper first treated the LTD algorithm as a special case of Tikhonov regularization that uses the approximation of the third-order derivative as the regularization factor. Then, to seek more flexible smoothness constraints, we successfully incorporated the smoothness seminorm used in variational interpolation theory into the reconstruction problem. Thus, the smoothing effects can be well understood according to the close relationship between the variational approach and the spline functions. Furthermore, employs two theories, i.e., Tikhonov regularization and spatial interpolation, to produce a smooth reconstruction by ORS-CT. Within the two theories’ frameworks, new other algorithms can be explored in order to improve the performance formulated by using different seminorms. For example, On the basis of this idea, we propose a new minimum curvature (MC) algorithm by based on the variational approach in the theory of the spatial interpolation using a seminorm approximating the sum of the squares of the curvature, which reduces the number of linear equations by to half comparing to that in the LTD algorithm using the biharmonic equation instead of the smoothness seminorm. We compared our The MC algorithm was compared with the non-negative least square (NNLS), GT-MG, and LTD algorithms by using multiple test maps. The MC and the LTD algorithms have The MC algorithm, compared with the LTD algorithm, shows similar performance as on-in terms of the reconstruction quality. But the MC algorithm but needs requires only aboutapproximately 65% the computation time of the LTD algorithm. It is much also simpler in realization than to perform than the GT-MG algorithm by using because it directly uses high-resolution grids directly during the reconstruction process to generate a high-resolution map immediately after one reconstruction process is done. Comparing
to-with the traditional NNLS algorithm, it shows better performance in three aspects: (1) the nearness of reconstructed maps is improved by more than 50%; (2) the peak location accuracy is improved by 1-2 m; and (3) the exposure error is improved by more than 10 times. The testing results show indicated the effectiveness of the new algorithm based on according to the spatial variational interpolation theory approach. More specific algorithms could be similarly further formulated and evaluated. Similarly, other algorithms may also be formulated to address problems such as the over-smooth issue in order to further improve the reconstruction equality. This study will promotes the practical application of the ORS-CT mapping of atmospheric chemicals.

1 Introduction

Measuring the concentration distribution of atmospheric chemicals over a large area is required in many environmental applications, including such as locating hot-spots or emission sources of air pollutants (Wu et al., 1999), understanding air pollutant dispersion and airflow patterns, and quantifying emission rates or ventilation efficiency (Samanta and Todd, 2000; Belotti et al., 2003; Arghand et al., 2015). The traditional network methods use multiple point samplers placed at various locations in the region under investigation, which This method is intrusive, time-consuming, and limited in temporal and spatial resolution (Cehlin, 2019). The advanced method is based on the combination of optical remote sensing (ORS) and computerized tomography (CT) techniques (ORS-CT). ORS-CT is a powerful technique for sensitive mapping of air contaminants measurement, which can detect a large throughout kilometer-size areas in situ and provide near-real-time information (Du et al., 2011). The path-integrated concentration (PIC) is measured along each path using techniques like Two commonly used ORS techniques use an open-path tunable diode laser (OP-TDL) or and open-path Fourier transform infrared spectrometer (OP-FTIR). The ORS analyzer emits a light beam targeted at multiple mirrors, which reflect the beam back to the analyzer. For each beam path, the path-integrated concentration (PIC) is obtained. After multiple PICs are collected, the two-dimensional concentration map can be reconstructed generated by through the CT technique tomographic reconstruction algorithms (Hashmonay et al., 2001). The ORS-CT method provides better spatial and temporal resolution than the network approach, and it is more sensitive than the range-resolved optical techniques. Comparing to other techniques, ORS-CT is also non-intrusive and fast. It also has good spatial resolution and can work suitable for continuously long-term and automatically monitoring. These advantages make ORS-CT have the potential to be an excellent tool for investigating air dispersion problems under various conditions.

In ORS-CT mapping of atmospheric chemicals, owing to factors including system cost, response time, beam configuration, In environmental applications, the number of beams in ORS-CT is usually less than 50 only tens, whereas the number of beams in medical CT is hundreds of thousands comparing to hundreds of thousands of beams used in the medical CT due to factors including cost, response, configuration, and other practical considerations. The very small beam number poses several
challenges in tomographic reconstruction algorithms. The reconstruction techniques have been well studied in medical CT. Theoretically, the true distribution can be determined exactly by Radon transformation approach, which requires infinite beams (Radon, 1986). In practice, the transformation methods approaches based on the theory of Radon transformation using a include back projection and filtered back projection (FBP) formula methods are not feasible because of noise and artifacts in the reconstructions (Radon, 1986; Herman, 2009). Series expansion based methods are another type of approaches which estimate Series expansion methods, which discretize the reconstruction problem before any mathematical analysis, are usually used in ORS-CT. The underlying distribution is represented by a linear combination of a finite set of basis functions (Censor, 1983), the true distribution by finding a finite set of numbers or superposition of a set of simple functions. In environmental applications, the number of beams in ORS-CT is usually less than 50 comparing to hundreds of thousands of beams used in the medical CT due to factors including cost, response, configuration, and other practical considerations. Therefore, only the series expansion based methods are applied in ORS-CT, which include the simplest type is the two main types of approaches: pixel-based approach and basis-function-based approach. The pixel-based approach, which divides an area into multiple grid pixels (grids) and assigns a concentration value to each pixel assigns a unit value inside each pixel. The PIC-path integral is approximated by calculated by summarizing the summation of the product of the pixel concentration value and the length of the path in each pixel. A system of linear equations can be set up for multiple beams. The inverse question problem is to finding the best-optimal set of pixel concentrations. Basis-function approach assumes that the true distribution is determined by the superposition of a set of simple basis functions with unknown parameters. Possible basis functions include bilinear function, bivariate Gaussian function, etc (Giuli et al., 1999; Hashmonay et al., 1999). A system of non-linear equations is set up for multiple beams. The question is to find the best set of parameters. For both approaches, the inverse questions are solved by minimizing the error function constructed based on according to some criteria (Price et al., 2001), including the least square criterion to minimize the summation of the squared errors between the observed and model-predicted PICs; the maximum likelihood (ML) criterion to maximize the probability of the PIC observations given the distribution of the random variables of the concentrations and observation errors; and the maximum entropy criterion to maximize the entropy of the reconstructed maps, given that the average concentration of the map is known including minimizing the L2-norm of error (finding a least-squares (LS) solution), maximum likelihood (ML), maximum entropy etc (Herman, 2009). In pixel-based reconstruction, the inverse problem is linear but usually ill-posed. The number of equations is very large. Commonly used numerical iterative techniques pixel-based algorithms are used to estimate the solution, including algebraic reconstruction techniques (ART) (ART), non-negative least square (NNLS) steepest descent, conjugate gradient (CG), and expectation-maximization (EM) (Tsui et al., 1991; Lawson and Janson, 1995; Todd and Ramachandran, 1994; Drescher et al., 1996). The NNLS algorithm has similar performance to the ART algorithm but shorter computation time (Hashmonay et al., 1999). It has been used in US EPA OTM-10 for horizontal radial plume mapping of air contaminants (EPA, 2005). The EM algorithm is mainly used for ML-based minimization. These traditional pixel-based algorithms are suitable for rapid CT, but they produce maps with poor spatial resolution, owing to the requirement that the pixel number must not exceed the beam number, or they
may have problem of indeterminacy associated with substantially underdetermined systems (Hashmonay, 2012). In the basis-function-based approach, the inverse problem is usually non-linear. Stochastic optimization algorithms need to be used to find the global minimum (e.g., simulated annealing (SA)).

To mitigate the problem of indeterminacy and improve the spatial resolution of reconstructions without substantially increasing the system cost, the smooth basis function minimization (SBFM) algorithm has been proposed. This algorithm represents the distribution map by a linear combination of several bivariate Gaussian functions (Drescher et al., 1996; Giuli et al., 1999). Each bivariate Gaussian has six unknown parameters (normalizing coefficient, correlation coefficient, peak locations and standard deviations) to be determined. The problem requires fitting these parameters to the observed PIC data. For ORS-CT mapping of atmospheric chemicals, this method iteratures show that the smooth basis function minimization (SBFM) algorithm performs better than other the traditional pixel-based algorithms (Drescher et al., 1996). This is because for ORS-CT applications because the patterns of air dispersion are physically smooth in shape (Wu and Chang, 2011). Therefore, an algorithm converging toward smooth concentration distribution consistent with the path-integrated data is a rational choice (Cehlin, 2019). However, the resultant equations defined by the PICs are non-linear because of the unknown parameters. The search for the best-fit set of parameters minimizing the mean-squared difference between predicted and measured path integrals can be performed through an iterative minimization procedure, such as the simplex method or simulated annealing.

The reported methods using simulated annealing to find a global minimization are SBFM is highly computationally intensive due to the non-linear optimization, which was reported to be about 100 times slower than pixel-based algorithms (Price et al., 2001). This makes it unfavorable thereby limiting the SBFM algorithm's practical applications, such as for rapid reconstruction, which is usually required in industrial applications such as monitoring chemical plants. However, an algorithm converging toward a smooth concentration distribution consistent with the path-integrated data has been demonstrated to be a rational choice. To improve the computational speed and append the smoothness a priori information to the inverse problem a representative pixel-based algorithm is the non-negative least square (NNLS), which is also the algorithm used in the USEPA OTM 10 for horizontal radial plume mapping (HRPM) measurements (EPA, 2005). It achieves similar results as the multiplicative algebraic reconstruction technique (MART), but with shorter computing time (Hashmonay et al., 1999). The traditional pixel-based algorithms, although suitable for rapid CT, have poor performance comparing to SBFM algorithm (Wu and Chang, 2011). This is mainly due to the coarse grid resolution limited by the beam number. Because the algorithm requires the number of grids to be less than or equal to the number of beams (Hashmonay, 2012).

To improve the performance of traditional pixel-based algorithms, there are two reported approaches used in ORS-CT. One method is the pixel-based 'low third derivative' (LTD) algorithm has been proposed, which This algorithm adds sets the third derivative at each pixel to zeros a smoothness restriction (Price et al., 2001)—thus resulting in a new system of linear equations that is overdetermined. The LTD algorithm has been reported to work nearly as well as the SBFM algorithm, but is about approximately 100 times faster (Price et al., 2001). Another method to produce the smoothness effect is the
'grid translation' (GT) algorithm, which shifts the basis grid by different distances (e.g., 1/3, or 2/3 the width of the basis grid) horizontally and vertically while keeping the basis grid fixed (Verkruysse and Todd, 2004). The performance is improved smoothly by averaging the reconstruction results after each shifting. There is also an improved version called 'grid-translation and multi-grid' (GT-MG), which applies the GT algorithm at different basis grid resolutions (Verkruysse and Todd, 2005). This method has been successfully used with the maximum likelihood expectation maximization (ML-EM) algorithm to improve the reconstruction accuracy, especially particularly in determining the peak location and value (Cehlin, 2019).

The successes of these algorithms show-demonstrates the necessary need of to applying the smoothness restriction to the ORS-CT gas mapping. Using With the LTD algorithm, we can easily achieve a smooth reconstruction is achieved by simply adding the third-order derivative constraints. The generated solutions are locally quadratic. However, like the Gaussian model used in the SBFM algorithm, the smoothness in the LTD algorithm is also achieved based on ideal model (quadratic functions) To. It is necessary to understand the characteristics of this these model constraints and in order to apply the algorithm method to specific application the theoretically basis of the LTD algorithm must be understood. However, this basis is was not clearly given defined in the literature. When we tried to With the purpose of introducing the smoothness as a priori information constraints to the reconstruction process, we found that the LTD algorithm can be treated as a special case of a smooth reconstruction could be achieved based on two well established theories. The first one is the Tikhonov regularization, a well-known technique to theory originated from Tikhonov for solving solve the ill-posed inverse problem (Tikhonov and Arsenin, 1977; Rudin et al., 1992). The most well known approach is the Tikhonov L2 regularization, which uses a penalty term defined by the squared norm of the ith-order derivative of the function as a regularization factor and produces a smoothing effect on the resulting solution (Gholami and Hosseini, 2013). From The third-order derivative is used in the view of regularization theory, the LTD algorithm is a regularized algorithm which uses the third order derivative. In fact, although the first, second, and higher order derivatives can also produce smooth results. A more flexible method of regularization uses the smoothness seminorm. The second one is the interpolation theory based on which almost all algorithms introduce the smooth effect to the output. Specifically, according to the variational interpolation approach theory, given its similar formula (Mitasova et al., 1995). The variational method is another way of achieving spline interpolation, given that the interpolation polynomial splines can be derived as the solution of certain variational problems of minimizing an integral whose integrant can produce a regularized inverse problem similar to Tikhonov regularization by using a smoothness seminorm. The seminorm consists of different order derivatives or their combinations. The solution to this inverse problem is a set of spline functions. For example, a bivariate smoothness seminorm with squares of second derivative leads to a thin plate spline (TPS) function. Based on the variational interpolation theory, the LTD algorithm is a special case of the regularized spline with tension (RST) (Mitasova et al., 1995).
The interpolation techniques are based on the given sample points, in contrast to tomographic reconstruction, in which only the line integrals are known. However, we have found that the interpolation can be adopted in the reconstruction process to produce a smooth solution by using the smoothness seminorm for interpolation as a smoothness regularization factor for the tomographic reconstruction problem. In the view of variational spline framework of the regularized inversion and spatial interpolation theories, the characteristics of the derivative-based algorithms have been well explored in the literature. The LTD algorithm can be considered as one case that minimizes the seminorm consisting of the third-order derivatives (Bini and Capovani, 1986). Other algorithms can also be formulated by using different seminorms. In this paper, we propose a new minimum curvature (MC) algorithm based on the variational interpolation using a seminorm approximating the integral of the squares of the curvature. This algorithm generates a smooth reconstruction approximating the application of cubic spline interpolation. We compared the algorithm with the NNLS, LTD, and GT-MG algorithms by using multiple test maps. We demonstrated its effectiveness and two main aspects of this method. First, smooth effect similar to spline interpolation is achieved during the reconstruction process by using high-resolution grid division, and second, it improves the computational efficiency is markedly better than that of comparing to the LTD algorithm by through using the corresponding biharmonic equation halving the number of linear equations instead of the according to the new smoothness seminorm to construct the additional linear equation at each pixel of the map, the new MC algorithm reduces the number of equations to half of the LTD algorithm and eventually reduces the computation time. This approach achieves the same performance but is easier to perform. Another innovation of the new algorithm is to use much larger number of grids than the number of beams instead of the comparable numbers of grids and beams in the traditional methods. Compared to the GT-MG algorithm which has the complicated operations involving multiple grids and grid translation operations in GT-MG algorithm. More specific algorithms applied for the ORS-CT method for mapping atmospheric chemicals could be further formulated and evaluated similarly, this approach offers the same performance but is much easier to realize. We compared the new MC algorithm with the NNLS, LTD, and GT-MG algorithms using multiple test maps. These tests showed the effectiveness of the new MC algorithm. This study also demonstrates the feasibility of introducing techniques from the Tikhonov regularization and spatial interpolation to the ORS-CT method for mapping atmospheric chemicals.

2 Materials and methodologies

2.1 ORS-CT and beam geometry

The area of the test field is 40 m× 40 m. We use the open-path tunable diode laser (OP-OPTDL) as the ORS analyzer, which is installed on a scanner and aimed at multiple retroreflectors by scanning periodically and continuously. The beam geometry can be categorized into overlapped and non-overlapped beam geometry based on the way
the beams are deployed. To compare with the results with those of the GT-MG algorithm, we used an overlapping beam configuration similar to the one that used by Verkruysse and Todd (2005).

As shown in Fig. 1, four TDL analyzers were located at the four corners of the test field. The retroreflectors were evenly distributed along the edges of the field. The total number of retroreflectors was 20. Each retroreflector reflected the laser beams coming from two different directions. Excluding the overlapped beams along the diagonals, the total beam number is 38. For the traditional pixel-based algorithm, the pixel number of grids should not exceed or equal to the beam number. Therefore, we divided the test field into $6 \times 6 = 36$ pixels. The concentration within each grid pixel is assumed to be uniform.

![Figure 1. The test field and beam configuration and grid division. (a) beam geometry; (b) beam geometry with the field was divided into $6 \times 6$ grid pixels. Four open-path TDL analyzers were located at the four corners. A total of 20 retroreflectors were distributed on the edges of the field.](image)

At each retroreflector, the path-integrated concentration (PIC was measured by the analyzer) is measured. The predicted PIC for one beam was equal to the sum of the multiplication of the pixel concentration and the length of the beam inside the pixel. In general, let us assume that the site is divided into $N_c = m \times n$ pixels, which are arranged as a vector according to the left-to-right and top-to-bottom sequence and indexed by $j$. The average concentration for the $j$-th pixel is $c_j$. The total number of laser beams is $N_b$, which are indexed by $i$. The length of the $i$-th laser beam passing the $j$-th pixel is $L_{ij}$.

Then, for the $i$-th beam, the measured PIC $b_i$ is contributed by all pixels. We have the following linear equation:

$$ pb_i = \sum_{j=1}^{N_c} L_{ij} c_j, \quad c_j \geq 0 \tag{1} $$

A system of linear equations can be set up for all beams

$$ pb = Lc, \quad c_j \geq 0 \tag{2} $$
where $L$ is the kernel matrix that incorporates the specific beam geometry with the pixel dimensions, $c$ is the unknown concentration vector of the pixels, and $b$ is a vector of the measured PIC data. With the least squares approach, the reconstruction can be solved by minimizing the following problem where $p$ is the PIC, $N_m x n$ is the total number of grids, $m$, $n$ - the row and column number of the grids, $i, j$ is the index of path number and cell number respectively, $L_w$ is the beam length of the $i$th beam in the $j$th cell, $c_j$ is the concentration of the $j$th cell.

$$
\min_{c} \|LC - b\|^2_2, \text{subject to } c \geq 0
$$

where $\| \cdot \|^2_2$ denotes the Euclidean norm. This non-negative constrained linear least squares inverse problem is can be solved by the widely used iterative NNLS optimization algorithm (NNLS) (Lawson and Janson, 1995), which is an active-set optimization method using an iterative procedure to converge on the best fit of positive values. The routine “lsqnonneg” in MATLAB software was used in this study. The optimal least squares solution is not smooth because the minimizing process does not introduce smoothness a priori information. Herein, the NNLS algorithm in the tomographic reconstruction refers to solving the original problem by using the NNLS optimization algorithm without adding additional a priori information. When the system of linear equations is underdetermined, the solution is not unique. Additional information must be introduced to choose the appropriate solution.

### 2.2 LTD algorithm and Tikhonov regularization and LTD algorithm

The LTD algorithm introduces the smoothness feature information by setting through setting the third-order derivatives of the concentration to be zero at each grid pixel in both $x$ and $y$ directions, which will thus generating solutions that are locally quadratic (Price et al., 2001). We define $c_i$ as an element of a one-dimensional (1-D) concentration vector of the pixels, but the pixels also have two-dimensional (2-D) structure according to the grid division of the site area and can be indexed by the row number $k$ and column number $l$, where $j=(k-1)n+l$. We use $C_{k,l}$ to denote the pixel concentration at the pixel located at the $k$-th row and $l$-th column of the grids. The third-derivative prior equations at the $(k, l)$ pixel are defined as Assuming the grid indices in $x$, $y$ directions are $i, j$, the new equations at one grid are:

\[
\begin{aligned}
\frac{d^3 C_k}{dx^3} &= (C_{k+2,l} - 3C_{k+1,l} + 3C_{k,l} - C_{k-1,l}) \frac{1}{\Delta x^3} (c_{k,l} + 3c_{k+1,l} - 3c_{k-1,l} + c_{k-2,l}) = 0 \\
\frac{d^3 C_k}{dy^3} &= (C_{k,l+2} \frac{1}{\Delta y} (c_{k,l+2} - 3C_{k+1,l} + 3C_{k-1,l} - C_{k-2,l}) = 0 \\
\end{aligned}
\]

(34)

where $\Delta d=\Delta x=\Delta y=\Delta d$ is the grid length in the $x$, $y$ direction divided in Eq. 3 to convert the grid-based derivatives into physical units. Therefore, two additional linear equations are introduced at each grid pixel defined by Eq. (34). There will be $2N_p$ linear equations appended to the original linear equations (defined by Eq. (2), thus resulting in a new over-determined system of linear equations with $(2N_p+N_i)$ equations and $N_p$ unknowns, that is over-determined.

A weight needs to be assigned to each equation depending on the uncertainty of the observation. Under the assumption that the analyzers have the same performance, the uncertainty is mainly associated with the path length. Therefore,
equations are assigned for the laser paths, weights are assigned inversely proportional to the path length to make sure that different paths have equal weight influences. In this application, the lengths of the laser paths are comparable (less than two times difference) approximately equal to each other. Thus, their weights are all set to the same value and scaled to be one. The weights for the third-derivative prior equations are assigned as the same value of $\mu$, because they are all based on the same grid length. The determination of $\mu$ follows the scheme for determining the regularization parameter described below. With the least squares approach, the reconstruction is intended to minimize the following problem:

$$\min_c \left\| L c - p \right\|_2^2 \quad \text{subject to } c \succeq 0$$

(5)

where $T$ is the kernel matrix for the third-derivative prior equations. Assuming that the new augmented kernel matrix is $A$ and the observation vector is $p$, the new system of linear equations will be $Ac = p$. The non-negative least squares solution can be found by traditional methods like the NNLS optimization algorithm. If the non-negative constraints are ignored, the least squares solution can be found analytically as $(ATWA)^{-1}ATWp$, where $W$ is a diagonal matrix whose diagonal elements are the weights (Price et al., 2001).

The LTD algorithm actually constructs a regularized inverse problem. It can be viewed as one special case of the Tikhonov $L_2$-regularization, which is the most well-known Tikhonov regularization technique. Its form can be written as the following minimization problem (Gholami and Hosseini, 2013):

$$\arg\min_{c \in \mathbb{R}} \min \|Lc - b\|_2^2 + \mu \|D_wc\|_2^2 + \mu \|R_e c\|_2^2$$

(4.6)

Where the first term represents the discrepancy between the measured and predicted values, the second term is the regularization term adding a smoothness penalty to the solution, $\mu$ is the regularization parameter controlling the conditioning of the problem, and matrix $R_e D_w$ is the regularization operator, which is typically an approximation of the $\ell_k$th-order difference derivative operator. The first- and the second-order difference derivative operators are commonly used.

We can see that for LTD algorithm, the uses the third-order derivative. The LTD algorithm uses the third-order derivative operator. Assuming the grid indices in $x$, $y$ directions are $i$, $j$, the third-order derivative at $y$-direction is

$$\frac{\partial^3 c}{\partial y^3} = c_{i,j+3} - 3c_{i,j+2} + 3c_{i,j+1} - c_{i,j}$$

(4)

The third-order difference forward difference operator in matrix form is

$$RD_3 = \begin{bmatrix}
-1 & 3 & -3 & 1 \\
-1 & 3 & -3 & 1 \\
\vdots & \ddots & \ddots & \vdots \\
1 & 3 & -3 & 1 \\
1 & 3 & -3 & 1
\end{bmatrix} \frac{1}{\Delta d} \in \mathbb{R}^{(m-3) \times n}$$

(5.7)
For pixel grids on the edges, we use the second-order derivative and first-order difference operators derivative can be used. The regularization parameter is set to be inversely proportional to the grid length. By setting the derivative to zero, the algorithm introduces two linear equations at each grid. The resulted system of linear equations is over-determined, which can be solved by NNLS algorithm. The regularization parameter is the same as the analogous to the weight assigned parameter for the prior equations to the third-derivative equations in the LTD algorithm. Therefore, \( \mu = w / \Delta d \), which is inversely proportional to the grid length.

The regularization parameter determines the balance between data fidelity and regularization terms. Determination of the optimum regularization parameter is an important step in the regularization method. It determines the balance between data fidelity and regularization terms. There are some parameter-choice methods existed with or without a priori information of the noise (Hamarik et al., 2012). However, the regularization parameter is problem and data dependent. There is no general-purpose parameter-choice algorithm which will always produce a good parameter. For simplicity, we use the commonly used method based on the discrepancy principle (Hamarik et al., 2012). The regularization parameter \( \mu (\mu_k) \) is chosen from a finite section of a monotonic sequence. For each value of \( \mu_k \), an optimal solution is derived by solving Eq. 4, the inverse problem. Then the discrepancy (the first term in Eq. 4) can be then calculated. The regularization parameter is determined to be the highest value that makes the discrepancy \( \|Lc - bp\|^2 = \|e\|^2 \) equal to \( \|e\|^2 = \text{the residual norm} N \sigma^2 \), where \( \sigma \) is the standard deviation of the noise which can be determined by examining the variance and uncertainty. In this application study, the reconstructions varied only slowly with the regularization parameters. Therefore, a precise selection of the weights parameter was not necessary. For computational efficiency, the regularization parameter can be selected from four widely varying values. The one produces the smallest discrepancy was used.

### 2.3 Variational approach interpolation and minimum curvature algorithm

Splines are special types of piecewise polynomials, which have been demonstrated to be very useful in numerical analysis and in many applications in science and engineering problems. They match given values at some points (called knots) and have continuous derivatives up to some order at these points (Champion et al., 2000). Spline interpolation is preferred over polynomial interpolation by fitting low-degree polynomials between each of the pairs of the data points instead of fitting a single high-degree polynomial. Normally, the spline functions can be found by solving a system of linear equations with unknown coefficients of the low-degree polynomials defined by the given boundary conditions.

The variational approach for interpolation provides a new way to find the interpolating splines and opens up directions in theoretical developments and new applications (Champion et al., 2000). Variational interpolation was motivated by the minimum curvature property of natural cubic splines, i.e., the interpolated surface minimizes an energy functional that corresponds to a physical bending energy. This principle provides flexibility in controlling the behavior of the generated spline. Given an observation \( z_k \) \( (k=1, \ldots, N) \) measured at the \( k \)-th point whose position vector is \( r_k \), a spline function \( F(r) \) for
interpolating the data points can be found through the variational approach by minimizing the sum of the deviation from
the measured points and the smoothness seminorm of the spline function the following problem
\[
\min_{c \in \mathbb{R}^{N_c}} \sum_{k=1}^{N} |F(r_k) - z_k|^2 + \omega I(F) \quad \{ \| Lc - p_i \|^2 + \omega I(c) \}
\]
(68)

where \( \omega \) is a positive weight, and \( I(F) \) denotes the square of smoothness seminorm. The seminorm can be defined in
various forms. The commonly used ones are the first, second, third derivatives, or their combinations. The solutions of to the
minimizing problems are spline functions, which can also be found by solving a Euler-Lagrange differential equation

We can see that the minimizing problem in Eq. (8) has a similar form to the Tikhonov regularization but with a more flexible
regularization term. The problem is that the variational interpolation is based on given data points, whereas the tomographic
reconstruction is based on measured line integrals. However, we show herein that the variational approach for interpolation
can also be applied to the latter problem to produce a smoothness solution with an effect similar to spline interpolation. In
addition, on the basis of different seminorms, we can formulate many different reconstruction algorithms. In this way, we
propose a new minimum curvature (MC) algorithm.

Under the assumption that the unknown concentration distribution is described by a function \( f(x, y) \), \((x_k, y_l)\) are the smallest
coordinates of the \( j \)-th pixel at row \( k \) and column \( l \) of the 2-D grids, then the concentration \( c_j \) equals the average concentration
of the pixel
\[
c_j = \frac{1}{(\Delta d)^2} \int_{x_k}^{x_{k+1}} \int_{y_l}^{y_{l+1}} f(x, y) dx dy
\]
(9)

The minimization problem according to the variational approach is formulated as
\[
\min_f \sum_{i=1}^{N_b} \sum_{j=1}^{N_c} |L_{ij} c_j - b_i|^2 + \omega I(f)
\]
(10)

For the MC algorithm, we define the seminorm. Based on different seminorms, we can formulate many different reconstruction
algorithms. As a demonstration, we propose the minimum curvature (MC) algorithm according to the minimum curvature
principle, which is used in geographic data interpolation to seek a 2-D surface with continuous second derivatives and
minimal total squared curvature (Briggs, 1974). The minimum-curvature surface is analogous to elastic plate flexure, and it
approximates the shape adopted by a thin plate flexed to pass through the observation data points with a minimum amount of
bending. This method generates the smoothest possible surface while attempting to follow the observation data as closely as
possible, which uses. The seminorm in the MC algorithm is equal to defined to be equal to the total square curvature:
\[
\| I(F) \|^2 = \int \int \left( \frac{\partial^2 c_f}{\partial x^2} + \frac{\partial^2 c_f}{\partial y^2} \right)^2 dx dy
\]
(711)

This integral must be discretized according to the grid division. According to the minimum curvature principle, the
minimization can be carried out by solving the biharmonic equation (Briggs, 1974)
\[
\frac{\partial^2 c}{\partial x^2} + 2 \frac{\partial^2 c}{\partial x \partial y} + \frac{\partial^2 c}{\partial y^2} = 0
\]
(8)
The discrete total square curvature is
\[ I = \sum_{k,l=1}^{M} \sum_{j,l=1}^{N} (I_{k,l,i,j}^2) \]
where \( I_{k,l,i,j} \) is the curvature at the \((k,l)\) pixel, which is a function of \( e_{C_{k,l,i,j}} \) and its neighboring grid pixel values. In two dimensions the approximation to the curvature is, in two dimensions, it is
\[ I_{k,l,i,j} = (eC_{k,l,i,j} + eC_{k-1,l,i,j} + eC_{k,l,i,j+1} + eC_{k,l,i,j-1} - 4eC_{k,l,i,j})/(\Delta d)^2 \]
To minimize the total squared curvature, we need
\[ \frac{\partial I}{\partial e_{C_{k,l,i,j}}} = 0 \]
Combining Eq. (911), (912), and (143), we get obtain the common following difference equation for the biharmonic equation
\[ [eC_{k,l,i+2,j} + eC_{k,i,j+2} + eC_{k,l,i,j-2}] \\
+2(eC_{k,l,i+1,j+1} + eC_{k-1,l,i,j+1} + eC_{k,l,i,j-1} + eC_{k,l-1,i,j-1}) \\
-8(eC_{k+1,l,j+1} + eC_{k-1,l,j-1} + eC_{k,i,j+1} + eC_{k,i,j-1}) + 20eC_{k,l,i,j})]/(\Delta d)^2 = 0 \]
This equation is appended at each grid pixel as a smoothness regularization. Therefore, there is only one additional prior equation at each grid. The seminorm can be calculated at each pixel using the finite difference approach. Each item in the summation is set to be zero to generate an equation at that pixel. Multiple items will lead to multiple equations at one pixel. This is how the LTD algorithm does to add additional equations. In this paper, however, we will only add one equation at each pixel to reduce the number of equations. This is done by using the corresponding Euler-Lagrange differential equation to the minimizing problem (Briggs, 1974). According to the minimum curvature principle, the minimization can be carried out by solving the biharmonic equation.
\[ \frac{\partial^2 e}{\partial x^2} + 2\frac{\partial^2 e}{\partial x \partial y} + \frac{\partial^2 e}{\partial y^2} = 0 \] (8)

The corresponding finite difference equation is
\[ e_{i+1,j} + e_{i-1,j} + e_{i,j+1} + e_{i,j-1} \\
+2(e_{i+1,j+1} + e_{i-1,j+1} + e_{i+1,j-1} + e_{i-1,j-1}) \\
-8(e_{i+1,j+1} + e_{i-1,j-1} + e_{i,j+1} + e_{i,j-1}) + 20e_{i,j} = 0 \] (9)
Therefore, one equation is set up for each grid instead of two equations in the LTD algorithm. For grid pixels on the edges, we set the approximation of the first and second derivatives to be zeros. Under the assumption that \( M \) is the kernel matrix of the prior equations, the reconstruction aims to minimize the following problem The weight \( \omega \) is set to be inversely proportional to the grid-length determined in the same way as determining the regularization parameter in the Tikhonov regularization method.
\[ \min_c \| Lc - b \|_2^2 + \lambda \| Mc \|_2^2, \text{ subject to } c \geq 0 \]  

where the parameter \( \lambda \) is determined in the same manner as the regularization parameter in Tikhonov regularization method. Similar to the LTD approach, the resulting constrained system of linear equations is over-determined and can be solved by the NNLS optimization algorithm.

Figure 2. Beam geometry and a with 30×30 grid division of the site grids.

For conventional pixel-based reconstruction algorithms, the number of grid pixels (unknowns) should not exceed the number of beams (equations) in order to get a well-posed system of linear equations. Because there are only dozens-tens of beams are usually used in an ORS-CT applications, the resultant grid-spatial resolution is very coarse. The GT algorithm is one way to increase the resolution because each translation uses a different grid division, and the high-resolution map is generated after each reconstruction process must be conducted for each grid division. In the MC algorithm, we use only one division of very-high-resolution grids directly before the reconstruction. The resultant system of linear equations remains determined because there is one of the derivative smoothness restriction at each pixel, the resulted system of linear equations is determined. As shown in Fig. 2, 30×30 grid pixels are used in the MC algorithm instead of the 6×6 grid pixels in the traditional NNLS algorithm approach. Under this configuration, the number of linear equations for the LTD algorithm is approximately 38+30×30×2=1838, whereas the number for the MC algorithm is approximately 38+30×30=938. Thus, the MC approach decreases the number of linear equations to approximately half that of the LTD algorithm. The smoothness feature seminorm of the MC algorithm will guarantee a smooth effect between all the pixels. This smoothing effect is similar to the spline interpolation applied after the reconstruction process, except that the interpolation...
achieved automatically when the inverse problem is solved. This aspect is important because an interpolation after the reconstruction cannot correct the error resulting from the reconstruction in terms of coarse spatial resolution. The MC approach evaluates the discrepancy based on the high-resolution values that are the same as the reconstruction outcomes. Errors due to coarse spatial resolution are corrected during the process.

2.4 Test concentration data

The NNLS, LTD, and MC algorithms were compared by using multiple test maps. The results were also compared with those of the GT-MG algorithm. In order to do this, we set up test conditions similar to those used in Verkruysse and Todd (2005).

The source-concentration distribution from one source is defined by a bivariate Gaussian distribution

\[ eg(x, y) = Q \exp\left(-\frac{(x-x_0)^2}{\sigma_x^2} + \frac{(y-y_0)^2}{\sigma_y^2}\right) \] (40)

where \( Q \) (0 to 40 ppm mg/m\(^3\)) is the source strength, \( x_0, y_0 \) (0 to 40 m) is the peak location, and \( \sigma_x, \sigma_y \) are the width of the peaks (with possible values of 2.8, 4.2, 5.7, and 7.1).

The source number varies from 1 to 5. For multiple sources, the resultant concentration distribution is the superposition value due to each source. For each source number, 100 maps were generated by randomly setting the source strength, location, and peak width from the defined ranges or set above.

2.5 Evaluation of reconstruction quality

A conventional image quality measure called nearness is used to describe the discrepancy between the original maps and the reconstructed maps. Nearness evaluates errors over all the grid cells in the map (Verkruysse and Todd, 2005)

\[ \text{Nearness} = \sqrt{\frac{\sum_{i=1}^{m} \sum_{j=1}^{n} (c_{i}^* - c_i)^2}{\sum_{i=1}^{m} \sum_{j=1}^{n} (c_{i}^* - c_{avg})^2}} \] (41)

where \( m, n \) are the grid divisions on the \( x, y \) direction of the map, \( c_{i}^* \) is the synthetic value of concentration in the \( i \)th grid on the map generated by the Gaussian distribution model, \( c_i \) is the estimated value for the \( i \)th grid in the map, and \( c_{avg}^* \) is the mean concentration of all the grids in the map. A nearness value of zero implies a perfect match.

The effectiveness of locating the emission source is evaluated by the peak location error, which calculates the distance between the true and reconstructed peak locations.

\[ \text{Peak location error} = \sqrt{(x_r - x_0)^2 + (y_r - y_0)^2} \] (42)

where \( x_r, y_r \) are the peak locations in the reconstruction map. For multiple peaks, only the location of the highest peak was calculated. The peak is located by searching for the largest concentration on the map. When multiple locations have the same values, the centroid of these locations is used.
Exposure error percentage is used to evaluate how well average concentrations in the whole field are reconstructed. It can reflect the accuracy of measuring chemical air emissions and emission rates from fugitive sources, such as agricultural sources and landfills (Verkruysse and Todd, 2004)

\[
\text{Exposure error \%} = \left| \frac{\sum_{i} c_{i} - \sum_{i} c_{i}^{\text{exact}}}{\sum_{i} c_{i}^{\text{exact}}} \right| \times 100\% \tag{431620}
\]

Herein, a measure using the resolution matrix is also applied to predict the reconstruction error due to different regularization approaches. Resolution matrices are commonly used to determine whether model parameters can be independently predicted or resolved, and how regularization limits reconstruction accuracy (Twynstra and Daun, 2012; von Clarmann et al., 2009).

Ignoring the non-negative constraints, the generalized inverse matrices for the NNLS, LTD, and MC algorithms can be found by

\[
\begin{align*}
G_{\text{NNLS}} &= (L^{T}L)^{-1}L^{T} \\
G_{\text{LTD}} &= (L^{T}L - \mu^{2}D_{3}^{T}D_{3})^{-1}L^{T} \\
G_{\text{MC}} &= (L^{T}L - \lambda^{2}M^{T}M)^{-1}L^{T}
\end{align*} \tag{21}
\]

The resolution matrix is defined as \( R = GL \). The reconstruction error is given by

\[
\delta c = c_{\text{model}} - c_{\text{exact}} = (R - I)c_{\text{exact}} - G\delta b \tag{22}
\]

where \( c_{\text{model}} \) and \( c_{\text{exact}} \) are the model-predicted and the exact concentrations, respectively, \( \delta b \) is the perturbation of the observation, \( I \) is the identity matrix, \( (R - I)c_{\text{exact}} \) is the regularization error caused by the inconsistency between the measurement data equations and the prior information equations, and \( G\delta b \) is the perturbation error.

For the LTD and MC approaches using high-resolution grids, the kernel matrix \( L \) is rank-deficient, and the regularized solution is robust to perturbation error over a wide range of regularization parameters. Thus, the perturbation error is negligible, and the reconstruction error is dominated by regularization error (Twynstra and Daun, 2012). Because the resolution matrix is determined only by the beam configuration and the regularization approach, it is independent of the actual concentration distribution. Therefore, it is best used to evaluate different beam configurations that considerably influence the reconstruction accuracy. However, in this study the beam configurations are fixed. We can therefore use the resolution matrix to measure different regularization approaches. In an ideal experiment, \( R = I \), thus implying that each unknown pixel value can be independently resolved from the measurement data. The regularization term forces the off-diagonal terms in \( R \) to be nonzero, thereby making the estimated concentration of each pixel a weighted average of the concentration of the surrounding pixels.

We can use the Frobenius distance between \( R \) and \( I \) defining a measure of fitness to predict the reconstruction error (Twynstra and Daun, 2012).

\[
\varepsilon = \frac{1}{N_{c}} \| R - I \|^{2}_{F} \tag{23}
\]
3 Results and discussions

In these tests, the traditional NNLS algorithm uses $6 \times 6$ grids, whereas the LTD and MC algorithms both use $30 \times 30$ grids. The results of the GT-MG algorithm are from Verkruysse and Todd (2005), in which they used a maximum basis grid resolution of $10 \times 10$ with $1/4$ grid size as translation distance was used. Of note, the test conditions were not exactly the same as those used by the GT-MG algorithm, which did not measure the peak location error and used a different method to calculate the exposure error by limiting the calculation domain to a small area near the peak instead of the entire map. Therefore, the results of the GT-MG algorithm are provided as a reference. Only the measure of nearness in the results of the GT-MG algorithm was compared because it did not give the peak location error, and the exposure error was calculated only in a small area near the peak instead of the whole map in this paper. The original resolution of the reconstruction map by the NNLS algorithm is too coarse (6.7 m). In order to determine the peak locations more accurately, all the reconstructed-concentration maps reconstructed by the NNLS algorithm were spline interpolated by spline function using with a resolution of 0.5 m. Fig. 3 depicts some examples of the test maps and reconstructed maps generated by different algorithms with different source numbers.

(a)

(b)

(c)
Fig. 3 depicts some examples of the test maps and reconstructed maps generated by different algorithms under different source numbers. As indicated by the nearness results, LTD and MC algorithms show better accuracy than the NNLS algorithm in the shapes and source strengths. Because the differences in accuracy (i.e., nearness, peak distance) between LTD and MC algorithms are very small, more specific evaluations may be needed to compare them using more complicated and realistic conditions.

3.1 Nearness

Table 1. Mean and standard deviation of nearness.

| Source number | NNLS   | LTD    | MC     | GT-MG* |
|---------------|--------|--------|--------|--------|
| 1             | 0.40 (0.21) | 0.13 (0.08) | 0.11 (0.07) | 0.09 (0.05) |
| 2             | 0.38 (0.16) | 0.15 (0.07) | 0.13 (0.06) | 0.16 (0.07) |
The nearness is the most important measure of accuracy of the reconstructed map. It represents the reconstruction of peak heights, shapes, and the production of artifacts. The smaller the number of sources nearness value, the better the reconstruction quality. In Table 1, the LTD, MC, and GT-MG algorithms generally reduce the nearness values by more than 50% with respect to the values obtained by the NNLS algorithm. Under the condition of one source, they can reduce the nearness by approximately about 70% with respect to the NNLS. The LTD, MC, and GT-MG algorithms show increasing trends as the source number increases, thus implying that the performance of the algorithm is affected by the complexity of the underlying distribution. The nearness results of NNLS for different numbers of sources are almost the same because they are the results after spline interpolation. In fact, the original un-interpolated results also show increasing trends. The interpolation improves the results of the NNLS algorithm more than those of the LTD and MC algorithms, which already use high-resolution grids. The overall performances of the LTD, MC, and GT-MG algorithms are very similar, whereas the new MC algorithm’s performances is slightly better.

### 3.2 Peak location error

Table 2. Mean and standard deviation of peak location error.

| Source number | NNLS (m)   | LTD (m) | MC (m) |
|---------------|------------|---------|--------|
| 1             | 1.78 (0.93)| 0.41 (0.45)| 0.40 (0.56) |
| 2             | 4.88 (8.21)| 1.97 (5.98)| 1.62 (4.81) |
| 3             | 5.17 (8.35)| 2.58 (6.77)| 2.34 (6.17) |
| 4             | 8.40 (11.53)| 5.22 (10.28)| 5.58 (10.76) |
| 5             | 8.95 (11.32)| 5.51 (10.15)| 5.77 (10.41) |

As shown in Table 2, the LTD and MC algorithms show better performance in peak location error than the NNLS algorithm. They generally improve the accuracy of peak location by 1 to 2 m. The errors of all the algorithms increase with the source number increase.

One reason for this finding is that because only the highest peak was calculated, a large error may happen when there are two or more peaks with that are comparable peak magnitudes on the map to each other in magnitude (see Fig. 3), the algorithm may not identify the correct location of the highest peak. Therefore, a large error may occur when the highest value on the reconstructed map is located on the wrong peak, this measure is more meaningful when the source number is small.
### 3.3 Exposure error

Table 3. Mean and standard deviation of exposure error.

| Source number | NNLS (%) | LTD (%) | MC (%) |
|---------------|----------|---------|--------|
| 1             | 29.26 (19.29) | 1.51 (2.11) | 1.61 (2.15) |
| 2             | 19.91 (11.99) | 1.05 (1.26) | 1.09 (1.19) |
| 3             | 16.05 (9.78)  | 1.16 (0.97) | 1.31 (0.92) |
| 4             | 12.99 (6.92)  | 1.15 (1.04) | 1.24 (1.00) |
| 5             | 12.13 (8.62)  | 1.30 (1.14) | 1.45 (1.16) |

As shown in Table 3, we can see a significant improvement in exposure error is achieved by the MC and LTD algorithms compared with the NNLS algorithm. The exposure error reflects the accuracy of the overall emissions measurement other than the concentration distribution. The performances of the LTD and MC algorithms are very similar to each other. Unlike the trends shown by the NNLS, it produces concentrations with very low spatial resolution and fits the true distribution better when the distribution becomes more uniform with larger numbers of sources. Because, implying that the NNLS algorithm uses coarse grid division, its performance in exposure error improves becomes better with the increasing of source number. A plausible cause of this phenomenon may be that the distribution becomes more uniform with larger numbers of sources. Because, implying that the NNLS algorithm uses coarse grid division, it produces concentrations with very low spatial resolution and fits the true distribution better when the distribution becomes more uniform is more suitable for relatively uniform distributions.

### 3.4 Computation time

Table 4. Mean and standard deviation of computation time.

| Source number | LTD (s) | MC (s) | Ratio (MC/LTD) |
|---------------|---------|--------|----------------|
| 1             | 11.08 (14.27) | 8.06 (10.03) | 0.73 |
| 2             | 21.02 (17.89) | 14.17 (11.46) | 0.67 |
| 3             | 34.44 (19.93) | 22.76 (13.61) | 0.66 |
| 4             | 43.58 (20.45) | 29.13 (12.92) | 0.67 |
| 5             | 59.48 (21.98) | 38.74 (15.05) | 0.65 |

In Table 4, the computation times for the LTD and MC algorithms are compared. The computation time generally increases with the increasing of source number. The MC algorithm is faster than the LTD algorithm because it has approximately about half the number of the linear equations than that in the LTD algorithm. The ratio result shows that the MC algorithm's computation time is approximately about 65% of that of the LTD algorithm when the source number is five. The trend of the ratio implies that the advantage of the MC algorithm becomes more obvious with the increasing complexity of the underlying distribution.
3.5 Fitness

Contour plots of the resolution matrices for the LTD and MC algorithms are shown in Fig. 4. (a) and (b). The fitness values for the LTD and MC algorithm are 1.4411 and 1.3878, respectively. The MC algorithm shows slightly better performance. The off-diagonal elements are not zeros. The reconstructed concentration at each pixel is a weighted average of the concentrations of the surrounding pixels according to the smoothness regularization. Each row of the resolution matrix can be regarded as smoothing weights. Because the pixels have a 2-D arrangement, we show the 2-D display of the row of the 106th pixel (row and column indices are 4 and 16) in the resolution matrix for the LTD and MC algorithms in Fig. 4. (c) and (d) as an example. The dependence on the beam geometry can be seen on both pictures. Because the beam configuration is fixed, the difference between the fitness values is mainly caused by the use of different regularization approaches. The fitness difference between the LTD and MC algorithms is very small, which may indicate that both algorithms have similar smoothness effects. This result coincides with the results from other measures discussed above. The 2-D display of the diagonal elements of the resolution matrix are shown in Fig. 4. (e) and (f), which are not much useful in this case.
3.6 Influence of the grid size

The derivatives are approximated by the finite differences during the discretization process. The finite grid length causes discretization error and affects the reconstruction results. We studied the influences of different grid divisions by investigating the changes of the nearness, peak location error, exposure error, and computation time with respect to the pixel number. Five different grid divisions were used: 6×6, 12×12, 18×18, 24×24, and 30×30. The peak number was five. A total of 100 maps were tested for each grid division. The results of the averaged values are shown in Fig. 5.
Figure 5. The change of (a) nearness (b) peak location error (c) exposure error percentage (d) computation time with respect to the pixel number.

The nearness, peak location error, and exposure error generally illustrate decreasing trends with increasing pixel number. The MC algorithm shows slightly better performance than the LTD algorithm with increasing pixel number. The performance improvement becomes slow for both algorithms when the division is finer than 24×24. The computation time shows approximately exponential growth trend with increasing pixel number. The LTD algorithm has a faster increasing rate than the MC algorithm. To conclude, the reconstruction performance is improved for both LTD and MC algorithms with increasing pixel numbers, but at the cost of exponential growth of the computation time. And the improvement becomes small when the resolution is higher than certain threshold value (24×24 herein). Therefore, there should be a balance between the performance and the computation time.

4 Conclusion

With the purpose of understanding the characteristics of the smoothness constraints and to seek more flexible achieving a smooth reconstruction, we first identified the LTD algorithm as a special case of the Tikhonov regularization. Then,
more flexible smoothness constraints were found through the smoothness seminorms and the according to variational spatial interpolation theory. The smoothness seminorms were successfully adopted can be applied to join the ORS-CT inverse problems. Based on the variational approach in the interpolation theory, we proposed a new MC algorithm by using a seminorm approximating the sum of the squares of the curvature the corresponding biharmonic equation instead of the smoothness seminorm to construct an additional linear equation at each pixel of the map. The new algorithm improves the computational efficiency comparing to the LTD algorithm through by reducing the number of linear equations by to half that of the LTD algorithm. It is simpler to perform than. Comparing to the GT-MG algorithm, which achieves high-resolution reconstruction by shifting the low-resolution base grids and averaging the resulted maps, the new MC algorithm uses by directly using high-resolution grids directly during the reconstruction process to generate a high-resolution map immediately after the reconstruction is done. Therefore, it is much simpler in realization.

The MC, LTD, and NNLS algorithms were compared by using multiple test maps with the GT-MG algorithm as a reference. The new MC algorithm shows almost the same similar performance as the LTD algorithm, but only needs requires about approximately 65% of its the computation time. The smoothness-related algorithms of LTD, MC, and GT-MG all show better performance than the traditional NNLS algorithm: the nearness of reconstructed maps is improved by more than 50%, the peak distance accuracy is improved by 1-2 m, and the exposure error is improved by more than ten 40 times. Because the differences in accuracy (i.e., nearness, peak distance) between the LTD and MC algorithms are very small, more specific evaluations may be needed to compare them by using more complicated and realistic conditions. These comparisons prove demonstrate the feasibility of introducing the theories of Tikhonov regularization and the theory of spatial variational interpolation techniques to mapping distribution of atmospheric chemicals using the ORS-CT techniques. With the basis of the seminorms these theories, it is easier to understand the advantages and the drawbacks of the current different algorithms. Common problems such as the over-smooth issue may be solved improved by testing formulating more algorithms suitable for techniques in these theories and using them in the ORS-CT applications. We need to note that although the smoothness is very good a priori information for the reconstruction problem, beam configuration and underlying concentration distribution are also important factors affecting the reconstruction quality. To further improve the reconstruction quality, extra a priori information based on the specific application may be added to the inverse problem. For example, the statistic information of the underlying distribution or the information resulting from the fluid mechanics.

Code and data availability. Data and code are available on request by contacting the authors.

Author contributions. KD was responsible for acquiring funding for this research. SL designed the algorithm and conducted the tests. SL and KD were both involved in data analysis. Both authors contributed to writing and editing the manuscript.
Competing interests. The authors declare that they have no conflict of interest.

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