Response to comments of reviewer 2

We thank the reviewer for the helpful comments and suggestions, and for careful reading of the manuscript. Listed below are our responses, with the original comment/question displayed in italics.

GENERAL COMMENTS

The paper describes a minimum curvature based regularization scheme for deriving 2-D trace gas concentrations from optical remote sensing and tomography. The chosen regularization scheme is sensible and the method seems to be an improvement over the state of the art in the field. The topic fits the journal.

The textual description is severely lacking and a rewrite to better guide the reader through the numerous methods is necessary. The description of the compared methods is severely lacking mathematical rigour and precise definitions causing the research to be not replicable in its current state.

I believe that the paper can only be published after a major revision and restructuring. See below for some general guidelines and a number of specific issues.

Thank you for the comments. We have done a major revision following your suggestions and addressed the issues. Please see the responses below for specific changes we have made to the manuscript.

MAJOR COMMENTS

Precise Mathematical description

The problem requires a much more precise mathematical introduction with clear definitions of employed terms. The paper gives a wide overview over several methods from literature and introduces many of these and related terms without clear definitions. At least those discussed later should be introduced well enough to follow the paper without further referencing.

The continuous and discrete view of the problem needs to be separated and the relationship clarified (see specific comments). Very often it is useful to specify the formulas for the continuous case and then “simply” discretize the resulting integrals and derivatives. In this case one achieves results that are
less dependent on the chosen discretization/gridding. This is particularly true as the sampling distance is (wrongly) used in the regularization strength instead of the integrals itself.

Thanks for the suggestions. We have redefined the symbols in this manuscript. Clear and precise descriptions are given. The continuous and discrete views have been distinguished by using different symbols. The issue with the sampling distance has been corrected. A new section of 3.6 has been added to discuss the influences of the grid size. Please refer to the supplement document for detailed description.

**Motivation for minimum curvature**

This chosen regularization is criticized for producing oversmoothened results. The major question is, what kind of regularization term would best describe the a priori information. The Laplacian is an obvious choice due to its relationship with the Poisson-Equation. If diffusion is the major process than a norm related to an exponential covariance would be very useful (see "Inverse Problem Theory" by Tarantola); also here, the Laplacian pops up at least for the 3-D exponential covariance. It would be interesting to motivate the choice of regularization form the underlying physics.

There is also a host of literature with respect to regularization for optical remote sensing methods from nadir and limb sounding satellites. It would be very interesting to put this method into this context and/or discuss the statistical angle.

It is true that the prior information should be based on the underlying physical process. The smoothness a priori information is most commonly used and very suitable for the diffusion process. More realistic prior information may be better for specific applications. For example, we have almost done investigating more complicated regularization based on the information derived from the air dispersion models for the diffusion-convection process. But the smoothness is the basic feature for all the problems in ORS-CT applications. In this study, we focus on better understanding the characteristics of this smoothness regularization. This is done by adopting the variational interpolation technique into the tomographic reconstruction process. Therefore, the smooth effect is closely related to the spline functions which has been well studied in the literature. Improving the performance of algorithms by using different seminorm is possible. The techniques used in similar field should be very inspiring. We are also expecting to explore more ideas from them. We also realize that currently the available algorithms for the 2-D tomographic mapping of the air contaminants are still very limited. This may be caused by the fact that the inversion problems are not exactly the same as those in similar area, especially when we look at the beam configurations and underlying distributions which considerably affect the performance of the algorithms. Therefore, algorithms need to be modified or adopted to be used in the specific applications. Overall, there are still lots of work to do in this area to promote the practical application of the ORS-CT technique.

**Diagnosis**

Due to the choice of a grid with more unknowns than measurements, diagnostics become more important. This can be done in a simple fashion with "resolution" measures. Rodgers' "Inverse Methods for Atmospheric Sounding" shows in great detail what kind of diagnostic quantities are relevant (resolution, measurement contribution, smoothing error, uncertainties...)

The nearness, peak location error, and the exposure error are the commonly used measures to evaluate the image quality. They provide direct measure of the performance only based on the final reconstruction results and independent of the reconstruction approaches. So they do not reveal the internal feature of the algorithms. In this view, the averaging kernel matrix is a good complementation.

According to the suggestion, we find that the averaging kernel matrix (resolution matrix) is commonly used in the atmospheric sounding but seldomly in the 2-D tomographic
reconstructions. In the latter case, it is largely affected by the beam geometry and is better to be used as a measure to evaluate the beam configuration. But it also reflects the regularization error given the same beam geometry in this study.

A new measure based on the resolution matrix have been added to the manuscript to determine whether the concentration can be independently predicted and how regularization limits reconstruction accuracy. The new fitness measure is defined as the average Frobenius distance between the resolution matrix and the identity matrix to predicate the reconstruction error. For the result, the MC algorithm shows slightly better performance than the LTD algorithm with a fitness value of 1.3878 comparing to 1.4411. The off-diagonal elements in the resolution matrix are not zeros. The reconstructed concentration at each pixel is a weighted average of the concentration of the surrounding pixels because of the smoothness regularization. Each row of the resolution matrix can be regarded as smoothing weights. The 2-D display of the row of the averaging matrix gives a clear dependence of the beam configuration.

Please refer to section 2.5, 3.5 in the supplement document for detailed description.

SPECIFIC COMMENTS

line 35
"which can detect a large area in situ and provide near real-time information" Maybe cover? Also "in situ" may be an unconventional use for a remote sensing instrument, depending on the community.

We have updated the description as following: "The ORS-CT technique provides a powerful tool for sensitive mapping of air contaminants over a kilometer-size area in real time."

line 43
To be precise both an infinite number of beams and beams of infinite length are required. One typically assumes a zero signal outside the reconstruction domain, which, for your problem, is a very reasonable assumption and at least alleviates the latter condition. How does the finite number of beams affect the solution?

It is a good idea to apply the zero-signal assumption outside of the domain, but in some applications of monitoring the air pollutants, this assumption may not be true. This is usually the case for mapping the concentration distribution over a horizontal plane because the plume may extend for a long distance outside of the mapping area, and the ORS-CT system cannot cover very large area due to the instrument limitation or deployment difficulties. We think this situation is different from the medical CT which reconstructs a slice with a clear boundary.

The number of beams affects the spatial resolution of the reconstructed map. The concentration of an area can be theoretically reconstructed if it is independently sampled by the beams. The beam number and geometry determine the size of an area that can be sampled independently. In the view of the reconstruction algorithm, the number of beams determines the number of equations of the inverse problem. The unknows are the concentration values of the grid pixels. To make the system of equations to be not ill-posed (underdetermined), it is required that the number of pixels should be no more than the number of beams. Otherwise, the problem of indeterminacy will occur and artifacts may appear on the resulted map. Because only tens of beams are used in ORS-CT, the resulted spatial resolution produced by the traditional pixel-based reconstruction algorithms is very coarse. This issue can be mitigated by adding the smoothness information to the inversion. We have rewritten the paragraph two in the introduction chapter to discuss the problem of the limited beam number.

line 44
"Series-expansion-based methods". Unclear what is meant here. Cite needed. The
explanation sounds like a simple discretization, transforming the "continuous problem" of finding a function over L2 to a discrete problem of identifying a number of samples. A reference has been added as suggested (Censor, 1983). The series expansion methods are distinguished from the transform methods by discretizing the problem at the very beginning whereas with transform methods the continuous problem is handled until the very end when the final formulas are discretized for computational implementation. In practice, the reconstruction domain is divided into discrete grid pixels. The pixel concentrations result from the linear combination of finite number of basis functions defined on the domain. We have updated the description in paragraph two.

line 48
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Also medical reconstruction techniques employ discrete samples and basis functions. With many more samples, obviously. The number of beams in the ORS-CT is very limited comparing to the medical CT (tens vs hundreds of thousands), which poses new challenges for the reconstruction techniques. The main issues are the coarse spatial resolution and the ill-posed inverse problem. For limited data tomographic reconstruction, the system of linear equations is rank-deficient. Additional a priori information like smoothness must be used to improve the performance. We have revised the texts in paragraph two to emphasis these challenges.

line 49
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I do not understand the difference between a pixel based approach and a basis function based. A pixel is a basis function with rectangular, non-overlapping support. The reviewer's understanding is accurate. There is ambiguity related to the classification of the methods. A pixel is one of the simplest basis functions with unit value inside the pixel and zero outside. The "basis function" in the manuscript is actually a "smooth basis function". To eliminate this ambiguity, we use only the "series expansion method", which is based on a linear combination of finite set of basis functions, as a category comparing to the transform method. And define the "pixel-based method" and "smooth basis function method" as two types of approaches using different basis functions. The text has been revised accordingly in paragraph two and three.

line 51
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"best" in what sense? We have updated the description as following text: "The inverse problem is to find the optimal set of pixel concentrations by minimizing the error function constructed based on some criteria, including minimizing the L2 norm of error (finding a least-squares solution), maximum likelihood (ML), maximum entropy, etc."

line 52
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Typically a basis is a basis of a (vector) space. Which space is spanned here? Is the full space spanned or only a subspace thereof? The two-dimensional reconstruction domain is divided into \( N=m \times n \) grid pixels. For the pixel-based methods, the space is the \( N \)-dimensional real space which is fully spanned by the \( N \) pixel-based basis functions. For smooth basis function methods, the space is the functions representing the distribution which may not be fully spanned depending on the choice of the basis functions. Taking SBFM algorithm as example, usually there are no more than five bivariate Gaussians are used so it can only represent some specific distributions.

line 53
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What parameters? Typically one derives pre-factors of normed basis functions.
Pre-factors are one type of parameters. But there may be more other parameters which need to be determined. For example, for the generalized Kaiser-Bessel window function, there are three parameters: sampling distance, window duration, and window shape. For the bivariate Gaussian in the SBFM algorithms, there are six parameters: normalizing coefficient, correlation coefficient, 2-D peak locations, and two standard deviations. But there are two ways dealing with these basis functions. First, the parameters are determined before the reconstruction according to mathematical analysis. Second, these parameters are kept unknown and the problem is to fit them to the observation data. The SBFM algorithm works in the latter way. We have revised the description of the SBFM algorithm to explain these parameters and how the algorithm solves the problem in paragraph three.

What equations? Why are those equations non-linear? Typically this problem would be linear even for non-trivial basis functions.
The equations are defined by the observed PIC data and the predicated values. The non-linear issue is specific for the SBFM algorithm, in which case the concentration values are calculated from linear combination of several bivariate Gaussians. But these Gaussians have unknown parameters as mentioned in the previous answer. The problem is to fit these parameters to the measurement data instead of getting the concentration values directly. Therefore, the bivariate Gaussians have to be calculated every time when a new set of parameters is proposed during the optimization process, which makes the problem non-linear and very computation intensive. We have revised the description of the SBFM algorithm to give more accurate explanation in paragraph three.

Best in what sense?
For the SBFM algorithm, the problem is to find the optimal set of parameters which best fits the observed PIC data by minimizing an error function using least square criterion. The text has been revised.

Too general. Fits to nearly any problem. What error function based on which criteria?
The least square criterion is to minimize the sum of the squared errors between the observed and model-predicted PICs. The maximum likelihood criterion is to maximize the probability of the PIC observations given the distribution of the errors. The maximum entropy criterion is to maximize the entropy of the reconstructed maps given the average concentration of the map is known. The manuscript has been updated accordingly.

Define ill-posed. Define very large (millions?).
The ill-posed problem refers to solve the underdetermined system of linear equation. For traditional approaches like ART and NNLS, the number of equations is determined by the number of beams which is usually hundreds of thousands for medical CT. But for ORS-CT, the number is only about tens. So "very large" is not accurate and has been deleted.

Many classes of non-linear problem can be solved efficiently by deterministic methods. Particularly convex optimization problems such as this.
It is true that the non-linear problems can be solved by deterministic methods. We have
revised the manuscript to make the description to be more accurate as following. First, the system of equations defined by the measured and predicated PICs are non-linear because of the presence of the bivariate Gaussians with unknown parameters. Second, the searching of the best-fit set parameters can be solved by iterative minimization procedure such as simplex method or simulated annealing. Third, simulated annealing method was used in the literature to find a global minimization when the cost function has many local minima. This minimizing process was computation intensive. It is possible to improve the speed by modifying the simulated annealing algorithm or using other optimization techniques. But currently, it is still very slow comparing to solving a system of linear equations.

**line 65**

*Exploiting previous (a priori) knowledge of a problem is almost always key in inverse problems. Doesn't dispersion/diffusion suggest a Laplacian as regularizing term? Is there a physical relationship between the dispersion processes and the minimum curvature? Yes. The Laplacian arise in the diffusion equation. Under steady state, the diffusion process is described by the Laplace's equation. In general case, the dispersion process is described by the convection-diffusion equation with additional convection and source/sink terms comparing to the diffusion process. The minimum curvature principle seeks a two-dimensional surface which minimizes the total squared curvature (the Laplacian power). The minimizing problem is also equivalent to solving a biharmonic equation. Therefore, we can investigate the similarity and difference between different algorithms from the equivalent equations. We have added more physical interpretation of the minimum curvature method in 2.3 section.*

**line 69**

*This is the first time NNLS is mentioned in the main text and a cite should be placed here with more detail. The EPA cite does not detail the NNLS algorithm. A reference has been added (Lawson and Janson, 1995).*

**line 74**

*If the number of unknowns is smaller than the number of measurements, such a problem may still be solved by using pseudo-inverses and or regularization techniques, which are computationally cheap.*

There are several things to mention here. It is true that an underdetermined linear system can be solved by pseudo-inverse method, which selects the solution with minimum Euclidean norm in this case. However, there is a problem of overfitting. This solution may not be physically realistic. It does not have the smoothness feature. Another thing is that the reconstruction is a constrained problem (non-negative solutions). The pseudo inverse may not generate a solution satisfying the constrains. Overall, the reconstruction of the underdetermined linear system is an ill-posed problem. Regularization techniques are needed to find an appropriate solution by adding additional realistic a priori information to the problem. For the ORS-CT reconstruction problem, the smoothness a priori information has been proven to be a good choice.

**line 88**

"But the theory basis of the LTD algorithm was not clearly given". By whom? *This paper is so far not helping in this regard.*

The LTD algorithm applies third-order derivative constrains to the solutions. But the underlying theory of this operation was not clearly defined in the literature. This prevents us from studying the characteristics of these constrains and getting a broad picture of the method. This manuscript first identifies the LTD method as one special case of the well-
established Tikhonov regularization. Then it studies more flexible smoothness constrains through the theory of variational interpolation, based on which the characteristic of different smoothness constrains are well understood through the close connection with the spline functions. Finally, we successfully adopted the variational approach from spatial interpolation problem into the tomographic reconstruction problem. We have rewritten the last two paragraphs in the introduction section to show these connections, and updated section 2.2 and 2.3 to give more description of the theories. Please refer to section 2.2, 2.3 in the supplement.

line 92
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What is regularization?
For the inverse or optimization problem, regularization is the process of adding information in order to solve an ill-posed problem or to prevent overfitting. The regularization term (penalty) imposes a cost on the optimization function to make the optimal solution unique.

line 95
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Interpolation theory typically deals within interpolation of (mostly discrete) data. How does that relate to the problem at hand?

This is one point that we want to show in this study. The interpolation techniques are based the given sample points, which is different from the tomographic reconstruction where only the line integrals are known. But we find that the variational interpolation technique can be adopted into the reconstruction process to produce a smooth solution. The connection is established based on two facts: (1) variational method is another way achieving the spline interpolation because the interpolating splines can be derived as the solution of certain variational problems of minimizing a seminorm defined by an integral consisting of different derivatives or their combinations; (2) this smoothness seminorm can also be used as a smoothness regularization factor for the tomographic reconstruction problem. In this way, the smooth effect similar to the spline interpolation can be achieved during the reconstruction process. We also find that other interpolation techniques can also be adopted in a similar way. We have rewritten the last paragraph of the introduction to describe this idea. Please also see section 2.3 in the supplement.

line 98
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"The solution to this problem is a set of spline functions." Please be precise about this. The algorithm derives, necessarily a vector. This vector can be interpreted in a various of ways. Of particular import is how it is interpreted by the "forward model", because that determines what is fit. This interpretation may differ from the interpretation for the regularisation term, but this introduces necessarily an error. One should be clear about that. Typically one sees the regularization term as an approximation: as the computation of derivatives by finite differences is inherently approximate. In the case that the gridding is very fine, the approximation error becomes small, and the point is moot, but this discussion is missing here. The discretization error has not been discussed and thus cannot be neglected. It is sensible to represent the 2-D field to be reconstructed here as a 2-D spline both in the forward model and the regularization term. This would remove approximation errors at the cost of a more complicated algorithms. Either way, the distinction and used assumptions must be made explicit and errors discussed.

Thanks for the good suggestions. The most important fact is that the variational approach is another way to find the interpolating splines. Therefore, by solving the minimization problem with the smoothness seminorm penalty, we will get a smooth solution similar to the effect of applying spline interpolation in the view of the forward model. Normally,
interpolation is based on the given data points, this study, however, successfully applied it
to the problem based on line integral data. The final solution is smooth and also has
characteristics related to the spline interpolation. But the "interpolation" is achieved during
the inversion process instead of after it, which is a key to improve the reconstruction
accuracy. We have rewritten the last two paragraphs in the introduction section to clarify
this relationship and the idea. Please also refer to the revised contents in section 2.3 in
the supplement for the description of the relationship between the variational interpolation
and spline function.

A new section of 3.6 has been added to discuss the discretization error due to the use of
different grid sizes. The changes of the nearness, peak location error, exposure error, and
computation time with respect to the pixel number were recorded using different grid
divisions. The results show that the nearness, peak location error, and exposure error
generally illustrate decreasing trends when increasing the pixel number. The MC algorithm
shows slightly better performance than the LTD algorithm with the increase the pixel
number. The performance improvements become slow for both algorithms when the
division is finer than 24 × 24. On the other hand, the computation time shows
approximately exponential growth trend with the increase of the pixel number. But the
LTD algorithm has a faster increasing rate than the MC algorithm. So there should be a
balance between the performance and the computation time. Please refer to section 3.6 in
the supplement for detailed description.

**line 102**

*Maybe a bit more of the theory should be described to make this more obvious to the reader.*

The Tikhonov regularization is a well-known technique to solve the ill-posed inverse
problem. The Tikhonov L2 regularization uses a penalty term defined by the squared norm
of the ith-order derivative of the function to produce a smoothing effect on the solution.
The variational interpolation is motivated by the minimum curvature property of natural
cubic splines. It is another way achieving the spline interpolation by the fact that the
interpolating polynomial splines can be derived as the solution of certain variational
problems of minimizing an integral consisting of different order derivatives or their
combinations. The interpolating data points can be found through minimizing the sum of
the deviation from the measured points and the smoothness seminorm. We have updated
the description in the last two paragraphs of the introduction and section 2.2 and 2.3 to
give more explanation of the Tikhonov regularization and the variational interpolation
theories. Please refer to section 2.2 and 2.3 in the supplement.

**line 104**

*Please properly and mathematically introduce the corresponding biharmonic equation and
the smoothness seminorm.*

The seminorm is defined as the total squared curvature of the concentrations in the MC
algorithm. The minimizing problem is equivalent to solving a corresponding biharmonic
equation. The definition of the biharmonic equation has been removed because we did not
use it for finding the solution in this study. The total squared curvature is first discretized.
Then in order to minimize the seminorm, we need its partial differential with respect to the
pixel concentration to be zero, which leads to a difference equation at each pixel. This
equation is appended at each pixel as a smoothness constrain. Thus, a new system of
liner equations is set up.

We have rewritten section 2.3 to mathematically define the smoothness seminorm. Please refer
to section 2.3 in the supplement for the formulas and description.

**line 106**

*Why does it half the number of equations?*
In the LTD algorithm, two equations are added at each pixel defined by the third-derivative prior information. While in the MC algorithm, only one equation is added that is derived from the minimizing of the total squared curvature. If we use 38 beams and a 30 x 30 grid division, there will be $38 + 30 \times 30 \times 2 = 1838$ equations for LTD, and $38 + 30 \times 30 = 938$ equations for MC. So the MC approach reduces the number of equations by approximately half. We have added the explanation in section 2.3. Please refer to it in the supplement.

**line 107**

*If the number of grid points increases what does that mean for the amount of information contained in the results and to what degree are the resulting "pixels" correlated? I.e. how well is the result resolved?*

We have added a new section 3.6 to show the influence of different grid sizes. The changes of the nearness, peak location error, exposure error, and computation time with respect to the pixel number were recorded using different grid divisions. The results show that the nearness, peak location error, and exposure error generally illustrate decreasing trends when increasing the pixel number. The MC algorithm shows slightly better performance than the LTD algorithm with the increase of the pixel number. The performance improvements become slow for both algorithms when the division is finer than 24 x 24. On the other hand, the computation time shows approximately exponential growth trends with the increase of the pixel number. But 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 the increase of the pixel numbers, but at the cost of exponential growth of the computation time. And the improvement become small when the resolution is higher than a certain threshold value (24 x 24 in this study). So there should be a balance between the performance and the computation time.

Please refer to section 3.6 for the detailed description.

**line 137**

*A very important interpretation of this approach is the statistical one (optimal estimation), where $R^TR$ can be interpreted as precision matrix codifying a priori information about the given distribution (i.e. smoothness). Please discuss.*

Thanks for the suggestion. A measure of fitness based on resolution matrix (averaging kernel matrix) has been used to investigate the regularization error caused by the inconsistency between the PIC equations and the prior information equations. This measure was defined as the average Frobenius distance between the resolution matrix and the identity matrix to predicate the reconstruction error.

A new section of 3.5 has been added to evaluate the results based on the fitness measure. The MC algorithm shows slightly better performance than the LTD algorithm with a fitness value of 1.3878 comparing to 1.4411. The off-diagonal elements are not zeros. The reconstructed concentration at each pixel is a weighted average of the concentration of the surrounding pixels because of the smoothness regularization. Each row of the resolution matrix can be regarded as smoothing weights. The 2-D display of the row of the averaging matrix gives a clear dependence of the beam configuration, while the diagonal elements may not provide much information in this case.

Please refer to section 2.5 and 3.5 in the supplement for the detailed description.

**line 139**

*In what sense is this an approximation? The given formula is discrete already, as such $R_i$ is not a derivative, but a finite difference operator.*

The approximation means the operator is not the actual derivative operator but an approximation of the derivative operator. We have changed them to “finite difference operator”.
The nomenclature is highly unusual. Typically derivatives are defined for continuous functions. c was so far a vector. This is an approximation of the third-order derivative of a function in y-direction by finite differences. And even then, the division by the grid-distance is missing. In this form, the regularisation is grid-dependent and would change in strength for different grid sizes, which requires a re-tuning of regularisation strength for every change of grid size. Please take the grid size into account.

Thanks for the correction. We have revised the symbol definitions in this manuscript. The discrete pixel concentrations can be arranged in both one-dimensional (1-D) and two-dimensional (2-D) forms. c is defined as a vector containing the concentrations in 1-D form with index of j. C is now a matrix containing the concentrations in 2-D form with row and column indices of k, l. The continuous distribution is described by a function of f(x,y). The division of the grid-distance has been moved to the finite difference equations. Therefore, the regularization parameter is independent of the grid size.

Please refer to section 2.2 and 2.3 in the supplement for the definitions.

Why is the regularization parameter set to 1 over grid length? To compensate for the missing factor in R_3, the power three is missing. There is a host of literature discussing optimal choice of this parameter (L-curve, optimal estimation, etc.). Practically, it is a tuning parameter which often requires manual adjustments unless both measurements and a priori are very well understood.

A weight needs to be assigned to each equation depending on the uncertainty of the observation. Assuming the analyzers have the same performance, the uncertainty is mainly related to the path length. Therefore, equations are assigned weights inversely proportional to path length to make sure different paths have equal influences. In this study, the lengths of the laser paths are approximately equal to each other. So their weights were all set to be the same value which is scaled to be 1. The weights for the third-derivative prior equations were assigned to be the same value of w because they were all based on the same grid length. The weight is analogous to the regularization parameter in the Tikhonov regularization and determined in the same way.

The regularization parameter determines the balance between data fidelity and regularization terms. Determination of optimum regularization parameter is an important step of the regularization method. 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 discrepancy principle. The regularization parameter is chosen from a finite section of a monotonic sequence. For each value of \( \mu \), an optimal solution is derived by solving the inverse problem. Then the discrepancy can be calculated. The regularization parameter is determined to be the highest value that makes the discrepancy equal to \( N_b \sigma^2 \), where \( N_b \) is the number of beams, \( \sigma \) is the standard deviation of the noise. In this study, the reconstructions varied only slowly with the regularization parameters. Therefore, precise selection of the parameter was not necessary. The regularization parameter was selected from four widely varying values. The one produced the smallest discrepancy was used.

We have added a separate paragraph to discuss how the weights and regularization parameter are determined at the end of section 2.3. Please refer to the description in the supplement.

What is meant by "setting the derivative to zero"? Formula (3) minimizes the expression and thus allows for non-zero derivatives unless the factor \( \mu \) is chosen to be very large.
The derivatives are set to be zero to generate constrain equations. That is how the third-derivative prior equations are set up and added to the original equations defined by the observations. That is true that the final optimal solutions may not have zero derivatives everywhere. They are approximately zero.

**line 152**

|c| is typically the absolute value of c. To describe more complicate regularization terms, one often uses a more general function \( \Phi(c) \) mapping \( \mathbb{R}^n \) to \( \mathbb{R}^+ \) or a norm with a subscription like \( ||c||_\phi^2 \). Are you referring to Sobolev-Norms?

Thanks for the suggestion. After revising the symbol definitions, c is a vector representing the discrete pixel concentrations. The seminorm is described by \( I(f) \) defined on the continuous distribution function of f. The discretization from of the seminorm is also provided in section 2.3 in the implement.

**line 155**

The solution to the problem is a discrete vector c, whereby each element of c defines the concentration in one pixel (see (1)). A spline is something very different, as it is a continuous. Your problem is set up to be non-continuous by definition. Please specify your model precisely and be careful with the distinction between the continuous and discrete view.

Thanks for the correction. We have distinguished the continuous and discrete view by using different symbols. Now this minimizing problem is defined in a continuous form. Please refer to section 2.1, 2.2 and 2.3 in the supplement for the detailed description.

**line 158**

c was defined as a vector, not as a continuous function.

Thanks for the correction. \( I(f) \) has been used to represent the seminorm defined on a continuous function f. Please refer to section 2.3 in the supplement for the revised definition.

**line 159**

What items in which summation? There is an integral in (7).

The discrete and continuous formulated have been distinguished. The discrete total squared curvature has been added. Please refer to Eq. (11) and (12) in section 2.3 in the supplement for the definitions.

**line 160:**

"This is how the LTD algorithm does to add additional(sic!) equations?" What does this mean?

Two additional linear equations are introduced at each pixel defined through the third-derivate prior information. There will be \( 2Nc \) linear equations appended to the original linear equations defined by the PIC observations, resulting in a new system of linear equations which has \( (2Nc + Nb) \) equations and \( Nc \) unknowns, where \( Nc \) is the pixel number, \( Nb \) is the beam number. Assuming the new augmented kernel matrix is \( A \), observation vector is \( p \), then the new system of linear equations is \( Ac=p \). We have revised the description in the section 2.3. Please see it in the supplement.

**line 161:**

Is such a complicated equation really more efficient computationally than two much simpler ones? What are the involved algorithmic complexities?
The LTD and MC algorithms use the same approach (the NNLS optimization algorithm) to solve the generated system of linear equations. The main different is the number of equations. The MC algorithm improves the computational efficiency by reducing the equation number by half comparing to the LTD algorithm.

**line 165:**

> It is unclear why this biharmonic equation is necessary. You are already minimizing a cost function in (6). Equation (7) gives you an immediate way to calculate $|c|$ required by (6). Computing discretized $ddc/ddx+ddc/ddy$ and computing the Euclidian norm should give you (6) without the need for higher derivatives in the definition of the problem. To efficiently solve (6) one might need higher derivatives depending on the chosen algorithm (e.g. Gauss-Newton), but your paper does not detail this part very well. Please describe in detail by which algorithm (6) is solved and how (9) plays a role in that.

The reviewer is correct. Solving the biharmonic equation gives the same solution to the minimization problem. But we did not find the solution in this way. Instead, we used the equations derived from the minimum curvature principle. To minimize the seminorm, the partial differential of the total squared curvature with respect to the pixel concentration needs to be zero for each pixel, resulting a difference equation. A new system of linear equations is set up based on these difference equations. The resulted inverse problem was solved by the NNLS optimization algorithm. We have removed the definition of the biharmonic equation. The detailed description of the derivation of the equations can be found in section 2.3 in the supplement.

**line 169:**

> Again, the regularization weight typically depends on diffusion coefficients and measurement errors and is often a tuning parameter. The grid size should directly be implemented in the finite difference equations.

The grid size has been implemented in the finite difference equations. A weight needs to be assigned to each equation depending on the uncertainty of the observation. Assuming the analyzers have the same performance, the uncertainty is mainly related to the path length. Therefore, equations are assigned weights inversely proportional to path length to make sure different paths have equal influences. In this study, the lengths of the laser paths are approximately equal to each other. So their weights were all set to be the same value which is scaled to be 1. The weights for the third-derivative prior equations were assigned to be the same value of $w$ because they were all based on the same grid lengths. $w$ was determined in the same way of determining the regularization parameter $\mu$ in the Tikhonov regularization.

The regularization parameter determines the balance between data fidelity and regularization terms. Determination of optimum regularization parameter is an important step of the regularization method. But 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 method based on discrepancy principle. The regularization parameter is chosen from a finite section of a monotonic sequence. For each value of $\mu$, an optimal solution is derived by solving the inverse problem. Then the discrepancy can be calculated. The regularization parameter is determined to be the highest value that makes the discrepancy equal to $N_b \sigma^2$, where $N_b$ is the number of beams, $\sigma$ is the standard deviation of the noise. In this study, the reconstructions varied only slowly with the regularization parameters. Therefore, precise selection of the parameter was not necessary. The regularization parameter was selected from four widely varying values. The one produced the smallest discrepancy was used. We have added a separate paragraph at the end of section 2.3 to discuss how the weights and regularization parameter are determined. Please refer to the supplement document.
What interpolation applied after the reconstruction process? The pixel-based algorithm assumes constant values over constant pixels. There is no smoothing interpolation, which would not deteriorate the fit to the measurements, i.e. deteriorate the results. What we want to explain here is that the variational approach is another way to find the interpolating splines. Therefore, by solving the minimization problem with the smoothness seminorm penalty, we will get a smooth solution similar to the result of applying spline interpolation in the view of the forward model. The final solution is smooth and also has characteristics related to the spline interpolation. But the "interpolation" is achieved during the inversion process instead of after it, which is a key to improve the reconstruction accuracy. This is important because an interpolation after the reconstruction cannot correct the error resulting from the reconstruction based on coarse spatial resolution. Please refer to section 2.3 in the supplement for revised description.

**line 186**

Here c is defined, for the first time, as a continuous function! Please properly distinguish the "different" c's. Thanks for the correction. g(x, y) has been used to represent the continuous concentration distribution.

**line 189**

What source number? (10) defines only a single source. If you use multiple sources, please accommodate this in (10). The source number varies from 1 to 5. For multiple sources, the resulted concentration distribution is the superposition due to each source. We have revised the description in section 2.4 to clarify this.

**line 189**

You state that the peak width was set randomly. Was it chosen randomly from the listed peak width of line 187f? Yes. The values were randomly chosen from the predefined set. We have updated the description to clarify this.

**line 195**

You were using c_i,j above for the 2-D fields, why now only c_i? The pixel concentration can be arranged both in 1-D and 2-D forms. We now distinguish these two arrangements by using c_j and C_k,l, where j is the index of a 1-D vector c, k, l is the row, column index of the 2-D matrix C respectively. The 1-D form is used for the linear equations as unknown vector. The 2-D form is used for the finite difference operations. Please refer to the section 2.2 in the supplement for the revised definitions.

**line 201**

How was the location of the highest peak located? The peak is located by searching the largest concentration value on the map. When there are multiple locations having the same values, the centroid of these locations is used. We have revised the manuscript to describe this.

**line 210**

Why did you not apply the other algorithm on your fields for better comparability? This study mainly focuses on the comparison between the new MC algorithm and the LTD
algorithm because they have similar formulas. While MG-GT algorithm uses a different approach. The detailed comparison with the MG-GT algorithm may be conducted in another study with the purpose to investigate their performance in different applications.

**line 215**
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While it seems to work, the pixel based algorithm derives pixels, not a continuous field. It is straightforward to derive a spline interpolated field directly, if desired for the higher accuracy. One simply needs to compute the integrals over the spline interpolated field for the coefficients when computing the error to the measurements. This can be accomplished by a linear matrix multiplication. I expect this to deliver similar results as the other methods at maybe even faster speed due to the smaller number of involved equations. Please discuss the choice of your simpler forward model.

With the purpose of achieving a smooth reconstruction, there is an important difference between applying interpolation after the reconstruction and applying smooth regularization during the reconstruction process. In the former situation, high-resolution grids cannot be used in order to make the inverse problem well-posed. The resulted solutions have coarse spatial resolution. Large error may occur and cannot be improved by the interpolation after the reconstruction has been done. In the latter case, high-resolution grids can be used during the reconstruction process. 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. Please refer to the revised explanation at the end of section 2.3 in the supplement.

**line 243**
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*Why does the necessary computation time scale with the number of sources? Shouldn't it be proportional to the problem size?*

It implies that the underlying distribution also affects the computation. Because when the source number is small, most of the area has almost zero values, which saves time to find an optimal solution. As the source number increases, the underlying concentration becomes complicated. It will need more iterations to approach an optimal solution. We have added a new section of 3.6 investigating the influence of the grid size. The result shows that the computation times for the LTD and MC algorithms exhibit approximately exponential growth trends with the increasing of the pixel number. While the LTD algorithm has a faster increasing rate than the MC algorithm. Please refer to section 3.6 in the supplement for the detailed discussion.

**line 283**
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*"oversmooth issue" - necessarily, the amount of information cannot increase between the measurements and the solution. Due to the chosen regularization, the result will be necessarily smooth. If it is "oversmooth" depends on whether the a priori assumption of smooth fields is correct or not.*

It is true that this issue depends on the actual underlying distribution. Actually, the underlying distribution has considerable influence on the performance of a reconstruction algorithm. In this study, we have adopted the variational interpolation approach to the tomographic reconstruction. As a result, now we can better understand the physical interpretation and the characteristics of applying the smooth regularization to the inversion according to the close connection with the spline functions. As a consequence, the issues related to the spline functions may also happen in the tomographic reconstructions and need to be further investigated.

In case that this assumption does not hold, "better" (less smooth) results can be achieved by Total-variation minimization (isotropic or anisotropic) and primal dual methods, e.g. Split-Bregman. I doubt this would fit better to your problem, though.

Actually, we have tried the TV minimization. But it did not give the smoothness effect we
expected. We think it is because the observations (line integrals) are too less to sufficiently distinguish the concentration gradient. As a result, an approach providing sharp gradient may give physically unrealistic solutions. However, this result is based on the specific synthetic problem in this manuscript. If different beam configurations (more beam number with optimized geometry) were used, the TV approach might be a good try.

MINOR REMARKS

line 55
"question". This is called an "inverse problem". corrected.

line 74
grids -> grid points?
The "pixel number" is used.

line 85
necessary -> necessity
"need" is used.

line 144
-> "third-order forward difference operator"
corrected.

line 160
Which "multiple items"?
This section has been rewritten and this sentence was removed.

line 174:
"For pixel-based"->"For conventional pixel-based", posted->posed corrected.