

Location Analysis of Observations for Atmospheric Chemical Transport Model with Emissions

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Abstract—Observations of the chemical states of the atmosphere typically have low temporal and spatial density. Based on the insufficient density of observations, one possibility to optimise the states estimation is to target the locations of observations which can potentially result in the largest forecast improvement. The objective of this work is to study the optimal locations of observations of atmospheric chemical transport model (CTM) with emissions by two approaches. One approach is the singular vector analysis, which can identify the sensitivity of observations by determining the directions of maximum perturbation growth per finite interval. Another approach is to transform the problem of optimal placement of observations to the duality of the linear-quadratic optimal problem of the locations of the control hardware, which, based on linear-quadratic optimal, can provide a global optimal solution for observation locations.

Index Terms—Chemical transport model, singular vector analysis, linear-quadratic optimal, observation locations, emission

I. INTRODUCTION

AIR quality and climate change are influenced by the fluxes of green house gases, reactive emissions and aerosols in the atmosphere. The chemical tendency with emissions in the atmosphere is usually described by CTM:

$$\frac{dx}{dt} = \mathcal{M}(x) + e(t)$$

where \mathcal{M} is a nonlinear model operator.

Further, the evolution of perturbation with the related tangent linear operator \mathbf{M}' is

$$\frac{d\delta x}{dt} = \mathbf{M}' \delta x + \delta e(t). \quad (1)$$

where $\delta x(t)$ is the perturbation evolved from the perturbation of initial states $\delta x(t_0) = x(t_0) - x^b(t_0)$ and emissions $\delta e(t) = e(t_0) - e^b(t)$, where $x^b(t_0)$ is the priori estimate of initial states and $e^b(t)$ is from the background knowledge of emissions.

However, the lack of ability to observe and estimate surface emission fluxes and important inner atmospheric fluxes with necessary accuracy is a major roadblock of hampering the progress in predictive skills of climate and atmospheric chemistry models. Obviously, based on our limited observations, the better choice of the locations of

observations can help us improving the predictive skills of atmospheric chemical states more efficiently. In this paper, two possible approaches, singular vector analysis and duality of problem of LQ optimal control location, are introduced to analyze the location of observations for general system, then we apply them into the atmospheric chemical transport model with emissions.

II. SINGULAR VECTOR ANALYSIS OF OBSERVATION LOCATION

Singular vector analysis (SVD) identifies the priorities of observations by detecting the fastest growing uncertainties. The targeted observations problem is an important topic in the field of numerical weather prediction. Singular vector analysis was firstly introduced to numerical weather prediction by Lorenz ([3]), who applied it to analyze the largest error growth rates in an idealised atmospheric model. Because of the high cost of computation, the singular vector analysis was not widely applied until 1980s. Then the method of singular vector analysis of realistic meteorological models with high dimension was possible ([2]).

In atmospheric chemistry, studies about observations are still not rich. Khattatov et al. ([8]) firstly analyzed the uncertainty of the chemical compositions. Liao et al. ([9]) focused on the optimal placement of observation locations of the chemical transport model. However, singular vector analysis for atmospheric chemistry with emissions is different since emissions play an similarly important role in forecast accuracy with initial values. Specified to this problem, Elbern et al. ([4]) provided a way to optimize initial values and emissions jointly by constant emission factors, based on 4-dimension variation approach. Goris et al. ([6]) recently used the singular vector decomposition to determine the sensitivity of the chemical composition to emissions.

For the study related to observations by singular vector analysis, we consider the linear continuous-time systems with the general integral solution:

$$\begin{aligned} \delta x(t) &= \mathbf{A}_{0,t} \delta x(t_0) \\ \delta y(t) &= \mathbf{C}_t \delta x(t) \end{aligned} \quad (2)$$

where the state space X is a separable Hilbert space and observation space Y is a finite-dimensional separable Hilbert spaces, $\mathbf{A}_{0,t} \in \mathcal{L}(X)$, $\mathbf{C}_t \in \mathcal{L}(X, Y)$.

We firstly define the magnitude of the perturbation of the initial state by the norm in X with a self-adjoint coercive operator A

$$\|\delta x(t_0)\|_A^2 = \langle \delta x(t_0), A \delta x(t_0) \rangle.$$

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Then, we define the magnitude of the related perturbation of observations $\delta y \in \mathcal{L}^2([t_0, t_N]; Y)$ for the entire fixed time interval $[t_0, t_N]$ by the norm

$$\|\delta y\|_{\{B_t\}}^2 = \int_{t_0}^{t_N} \langle \delta y(t), B_t \delta y(t) \rangle dt,$$

where $\{B_t\}$ is a set of positive operators.

The ratio of the magnitude of observation perturbation and the initial perturbation can be written as

$$\begin{aligned} \sigma^2 &= \frac{\|\delta y\|_{\{B_t\}}^2}{\|\delta x(t_0)\|_A^2} = \frac{\int_{t_0}^{t_N} \langle \delta y(t), B_t \delta y(t) \rangle dt}{\langle \delta x(t_0), A \delta x(t_0) \rangle} \\ &= \frac{\int_{t_0}^{t_N} \langle \mathbf{C}_t \delta x(t), B_t \mathbf{C}_t \delta x(t) \rangle dt}{\langle \delta x(t_0), A \delta x(t_0) \rangle} \\ &= \frac{\int_{t_0}^{t_N} \langle \delta x(t), \mathbf{C}_t^* B_t \mathbf{C}_t \delta x(t) \rangle dt}{\langle \delta x(t_0), A \delta x(t_0) \rangle} \\ &= \frac{\int_{t_0}^{t_N} \langle \delta x(t_0), \mathbf{A}_{0,t}^* \mathbf{C}_t^* B_t \mathbf{C}_t \mathbf{A}_{0,t} \delta x(t_0) \rangle dt}{\langle \delta x(t_0), A \delta x(t_0) \rangle} \\ &= \frac{\langle \delta x(t_0), (\int_{t_0}^{t_N} \mathbf{A}_{0,t}^* \mathbf{C}_t^* B_t \mathbf{C}_t \mathbf{A}_{0,t} dt) \delta x(t_0) \rangle}{\langle \delta x(t_0), A \delta x(t_0) \rangle} \end{aligned} \quad (3)$$

Denote the integral operator in (4) by

$$\mathcal{G} = \mathcal{C}^* \mathcal{C} = \int_{t_0}^{t_N} \mathbf{A}_{0,t}^* \mathbf{C}_t^* B_t \mathbf{C}_t \mathbf{A}_{0,t} dt, \quad (5)$$

where $\mathcal{C} : X \rightarrow \mathcal{L}^2([t_0, t_N]; Y)$ is the observability map defined by

$$\mathcal{C}x(t_0) := B_t^{\frac{1}{2}} \mathbf{C}_t \mathbf{A}_{0,t} x(t_0), \quad x(t_0) \in X$$

and its adjoint operator \mathcal{C}^* is

$$\mathcal{C}^* y = \int_{t_0}^{t_N} \mathbf{A}_{0,t}^* \mathbf{C}_t^* B_t^{\frac{1}{2}} y(s) ds.$$

For searching the different directions of error growth and reconstructing δy from the initial perturbation, we need to find the following eigenvalues and eigenvectors

$$\begin{aligned} A^{-\frac{1}{2}} \mathcal{C}^* \mathcal{C} A^{-\frac{1}{2}} v_k(t_0) &= \sigma_k^2 v_k(t_0) \\ \mathcal{C} A^{-\frac{1}{2}} A^{-\frac{1}{2}} \mathcal{C}^* u_k(t) &= \sigma_k^2 u_k(t) \end{aligned}$$

then, we have

$$\delta y(t) = \sum_{i=1}^{\infty} \sigma_i \langle \delta x(t_0), v_i(t_0) \rangle u_i(t) \quad (6)$$

Specified to observation location analysis, we let \mathbf{L} be the location projection operator satisfying:

$$\mathbf{L}(s) = \begin{cases} 1, & s \in L, \\ 0, & \text{otherwise.} \end{cases}$$

where L denotes the set of observation locations.

Hence, considering the norm of $\mathbf{L} \delta y$ instead of δy in (3) and from (6), it is clear that δy is sensitive to the initial perturbation if the coefficients related to large singular values are dominant. Equivalently, it means the related observation locations are efficient for improving the estimate of initial states.

Now, let us consider the stochastic finite-dimensional system for application and take the Kalman filter into account. For system

$$\delta x(t_{k+1}) = \mathbf{A}_{t_k, t_{k+1}} \delta x(t_k) \quad (7)$$

$$\delta y(t_k) = \mathbf{C}_{t_k} \delta x(t_k) + \nu_{t_k}, \quad \nu_{t_k} \sim N(0, R_k),$$

according to discrete-time Kalman filter, we have

$$P_{k|k}^{-1} = P_{k|k-1}^{-1} + \mathbf{C}_{t_k}^T R_k^{-1} \mathbf{C}_{t_k},$$

$$P_{k+1|k} = \mathbf{A}_{t_k, t_{k+1}} P_{k|k} \mathbf{A}_{t_k, t_{k+1}}^T,$$

where $P_{k+1|k}$ is the forecasting covariance of the states for the time step $k+1$, $P_{k|k}$ is the analysing covariance of the states at time step k , and $\mathbf{A}_{t_k, t_{k+1}}$ is the evolution matrix from time k to $k+1$.

Hence, by iteration, we have

$$\begin{aligned} P_{k+1|k}^{-1} &= \mathbf{A}_{t_0, t_{k+1}}^{-T} P_{0|-1}^{-1} \mathbf{A}_{t_0, t_{k+1}}^{-1} \\ &\quad + \sum_{i=0}^k \mathbf{A}_{t_i, t_{k+1}}^{-T} \mathbf{C}_{t_i}^T R_i^{-1} \mathbf{C}_{t_i} \mathbf{A}_{t_i, t_{k+1}}^{-1}, \end{aligned}$$

then,

$$\begin{aligned} P_{0|k}^{-1} &= \mathbf{A}_{t_0, t_{k+1}}^T P_{k+1|k}^{-1} \mathbf{A}_{t_0, t_{k+1}} \\ &= P_{0|-1}^{-1} + \sum_{i=0}^k \mathbf{A}_{t_0, t_i}^T \mathbf{C}_{t_i}^T R_i^{-1} \mathbf{C}_{t_i} \mathbf{A}_{t_0, t_i}, \end{aligned}$$

where $P_{0|k}$ can be viewed as the analysing covariance of the initial states obtained from $P_{k+1|k}$, $P_{0|-1}$ is the first guess of initial covariance.

Further, we substitute B_t in (5) to R_t and restrict the system in the fixed time interval $[t_0, \dots, t_N]$

$$P_{0|N}^{-1} = P_{0|-1}^{-1} + \mathcal{C}^* \mathcal{C}.$$

For measuring the improvement of the estimation with the initial guess $P_{0|-1}$, we define the improvement covariance as

$$\begin{aligned} &P_{0|-1}^{-\frac{1}{2}} (P_{0|-1} - P_{0|N}) P_{0|-1}^{-\frac{1}{2}} \\ &= P_{0|-1}^{-\frac{1}{2}} (P_{0|-1} - (P_{0|-1}^{-1} + \mathcal{C}^* \mathcal{C})^{-1}) P_{0|-1}^{-\frac{1}{2}} \\ &= I - (I + P_{0|-1}^{\frac{1}{2}} \mathcal{C}^* \mathcal{C} P_{0|-1}^{\frac{1}{2}})^{-1}. \end{aligned} \quad (8)$$

Assuming $A = P_{0|-1}^{-1}$ and representing (8) by matrices, we have

$$P_{0|-1}^{\frac{1}{2}} \mathcal{C}^* \mathcal{C} P_{0|-1}^{\frac{1}{2}} = V \Sigma^2 V^T,$$

where V is a unitary matrix consisted by the singular vectors, Σ is the diagonal matrix consisted by the singular values.

From (8),

$$\begin{aligned} &P_{0|-1}^{-\frac{1}{2}} (P_{0|-1} - P_{0|N}) P_{0|-1}^{-\frac{1}{2}} \\ &= I - (I + V \Sigma^2 V^T)^{-1} \\ &= V V^T - (V V^T + V \Sigma^2 V^T)^{-1} \\ &= V (I - (I + \Sigma^2)^{-1}) V^T \\ &= \sum_{i=1}^{2n} \frac{\sigma_i^2}{1 + \sigma_i^2} v_i v_i^T. \end{aligned}$$

Define

$$\tilde{P} = P_{0|-1}^{-\frac{1}{2}}(P_{0|-1} - P_{0|N})P_{0|-1}^{-\frac{1}{2}}$$

and consider the nuclear norm for matrices, we obtain

$$\|\tilde{P}\|_1 = \sum_{i=1}^n \frac{\sigma_i^2}{1 + \sigma_i^2} \text{tr}(v_i v_i^T).$$

If the norm of \tilde{P} is very close to zero, this means the observations cannot indeed help us improving the estimation of emissions, otherwise observations is of high efficiency to improve the estimation.

Especially for CTM with emissions, it is novel in our current work to establish the model with a new combination of the initial values and emissions, which can be showed as follows:

Firstly, because emissions can be estimated by constant emission factors ([4]), it is reasonable to construct the evolution of emissions as

$$e(t) = \mathbf{E}_t^b e(t_0), \quad (9)$$

where \mathbf{E}_t^b is from the inaccurate background knowledge of emissions, satisfying

$$e^b(t) = \mathbf{E}_t^b e^b(t_0).$$

Secondly, from (1) with the integral operators $\mathbf{M}_{0,t}$ and $\mathbf{M}_{0,t}^e$ respectively for states and emissions we have

$$\delta x(t) = \mathbf{M}_{0,t} \delta x(t_0) + \mathbf{M}_{0,t}^e \delta e(t_0). \quad (10)$$

Then, consider (9) and (10) together, we obtain

$$\begin{pmatrix} \delta x(t) \\ \delta e(t) \end{pmatrix} = \begin{pmatrix} \mathbf{M}_{0,t} & \mathbf{M}_{0,t}^e \\ 0 & \mathbf{E}_t^b \end{pmatrix} \begin{pmatrix} \delta x(t_0) \\ \delta e(t_0) \end{pmatrix}. \quad (11)$$

(11) provides the possibility for improving the estimation of both initial values and emissions by Ensemble Kalman smoother, which not only can optimize the initial values and emissions but also can update their weights.

Further, it is also possible to analyse the sensitivity of observation locations for (11) by SVD. Meanwhile, the improvement of estimation also can be measured by nuclear norm by SVD according to the above analysis.

III. PROBLEM OF LQ OPTIMAL OBSERVATION LOCATION

From the above discussion, it is clear that singular vector analysis can provide the sensitivity of observation locations to the initial perturbation, but we cannot obtain an optimal solution by this approach. Hence, in this section, we mainly focus on the optimal problem of observation location.

It is well known that the choice of the placement of control hardware, such as sensors and actuators, plays an important role in the designs of control systems. Many researchers have focused on the study of finding the optimal locations of control hardwares and different criteria of optimising control location were established, such as maximization of observability and controllability ([7], [14]), minimizing the linear quadratic regulator cost ([13]). Geromel ([5]) successfully reformulated the LQ cost function into a convex optimization problem by mapping the locations of controller

into zero-one vectors and expressed the solution of classic LQ problem in terms of the Riccati Equation. Morris ([12]) optimized the controller location by the norm of a matrix P over possible controller locations which can be obtained by solving an algebraic Riccati equation. Further, Darivandi et al. ([3]) provided an algorithm for the above LQ optimal problem of controller location.

However, the objective of our work is to optimize the location of observations for CTM with emissions. Though the importance of observations is obvious for determining the predictive skill of a dynamic system, by authors' knowledge, few attempts have been made at solving this problem, especially based on LQ optimization method. Therefore, for the general linear stochastic system

$$\begin{aligned} x(t) &= T(t, t_0)x(t_0) + \int_{t_0}^t T(t, s)[B(s)u(s) + Q(s)\omega(s)]ds, \\ y(t) &= C(t)x(t) + D(t)v(t) \end{aligned}$$

where \mathcal{X} , \mathcal{Y} and \mathcal{E} are seperable real Hilbert spaces respectively with the Borel fields \mathcal{B} , \mathcal{B}_Y , \mathcal{B}_E and the Gaussian measures μ , μ_y and $\mu_e \sim N(0, I)$, $x(t)$, $y(t)$, $\omega(t)$ and $v(t)$ are the random variables in \mathcal{X} , \mathcal{Y} and \mathcal{E} , $T(\cdot, \cdot)$ is a uniform bounded mild evolution operator on \mathcal{X} , \mathcal{U} is also a seperable real Hilbert space with $B \in L_{s,\infty}(t_0, t_f; \mathcal{U}, \mathcal{X})$, $B^* \in L_{s,\infty}(t_0, t_f; \mathcal{X}, \mathcal{U})$ and $u \in L_2([t_0, t_f]; \mathcal{U})$, $C \in L_{s,\infty}(t_0, t_f; \mathcal{X}, \mathcal{Y})$, $D \in L_{s,\infty}(t_0, t_f; \mathcal{E}, \mathcal{Y})$ and DD^* is coercive, $Q \in L_{s,\infty}(t_0, t_f; \mathcal{E}, \mathcal{X})$. where $L_{s,\infty}(t_0, t_f; \mathcal{X}, \mathcal{Y}) \triangleq \{F : [t_0, t_f] \rightarrow \mathcal{L}(X, Y) | F \text{ is strong measurable and } \|F\|_\infty := \text{ess sup}_{t \in [t_0, t_f]} \|F(t)\| < \infty\}$.

In Hilbert spaces, without the differentiability of $T(\cdot, \cdot)$, we can also conclude that for the filter problem, the best unbiased estimation $\hat{x}(t)$ of $x(t)$ has the form

$$\begin{aligned} \hat{x}(t) &= T_L(t, t_0)\hat{x}(t_0) + \int_{t_0}^t T_L(t, s) \cdot \\ &\quad [P(s)C^*(s)(D(s)D^*(s))^{-1}y(s) + B(s)u(s)]ds \end{aligned}$$

where $L = PC^*(DD^*)^{-1}$,

$$T_L(t, t_0)x = T(t, t_0)x - \int_{t_0}^t T_L(t, s)L(s)C(s)T(s, t_0)x ds.$$

and its covariance is

$$\begin{aligned} P(t) &= T_L(t, t_0)P(t_0)T_L^*(t, t_0) + \int_{t_0}^t T_L(t, s) \\ &\quad [Q(s)Q^*(s) + L(s)D(s)D^*(s)L^*(s)]T_L^*(t, s)ds. \end{aligned}$$

It can be shown the nuclear norm of $P(t)$ can be considered as the measurement of the estimation error.

By observing the similarity of the filter problem and the linear quadratic optimal control, it is clear that considering the above covariance $P(t)$ is equivalent to consider the Riccati operator $\Pi(t_f - t)$ which satisfies the minimal solution of the cost functional as follows

$$\langle x(t), \Pi(t)x(t) \rangle = \min_{u \in L_2([t, t_f]; U)} J(t, x(t), u),$$

where

$$J(t, x(t), u) = \langle x(t_f), P(t_0)x(t_f) \rangle + \int_t^{t_f} \langle Q^*(t_f - s)x(s), Q^*(t_f - s)x(s) \rangle + \langle u(s), D(t_f - s)D^*(t_f - s)u(s) \rangle ds,$$

and

$$x(t) = T^*(t_f, t_f - t)x(t_0) + \int_{t_0}^t T^*(t_f, t_f - s)C^*(t_f - s)u(s)ds.$$

Due to the duality of filter problems and linear quadratic optimal control, it is sufficient to focus on the study of location control of linear quadratic problem.

Now consider the situation that we have the opportunity to choose m locations to control and each location varies over a compact set $\Omega \in \mathbb{R}^l$. We denote the m locations by the parameter r . Obviously, $r \in \Omega^m$. Then the time-varying system with location-dependent control operator B_r is

$$x(t) = T(t, t_0)x(t_0) + \int_{t_0}^t T(t, s)B_r(s)u(s)ds, t_0 \leq t \leq t_f \quad (12)$$

with the cost functional

$$J_r(t, x(t), u) = \langle x(t_f), Gx(t_f) \rangle + \int_t^{t_f} \langle C(s)x(s), C(s)x(s) \rangle + \langle u(s), R(s)u(s) \rangle ds \quad (13)$$

then according to the well-known results of linear quadratic control, we have

$$\min_{u \in L_2([t, t_f]; U)} J(t, x(t), u) = \langle x(t), \Pi_r(t)x(t) \rangle, t \in [t_0, t_f],$$

with the optimal control

$$u(t) = -K_r(t)x(t) = -R(t)^{-1}B_r^*(t)\Pi_r(t)x(t),$$

and the optimal trajectory

$$x(t) = T_{K,r}(t, \tau)x(\tau),$$

where

$$T_{K,r}(t, \tau)x = T(t, \tau)x - \int_{\tau}^t T_{K,r}(t, s)B_r(s) \cdot R^{-1}(s)B_r^*(s)\Pi_r(s)T(s, \tau)x ds.$$

and the self-adjoint operator $\Pi_r(t)$ satisfies the integral Riccati equation

$$\begin{aligned} \Pi(t) &= T_{K,r}^*(t_f, t)GT_{K,r}(t_f, t) \\ &+ \int_t^{t_f} T_{K,r}^*(s, t)[C^*(s)C(s) \\ &+ \Pi_r(s)B_r(s)R^{-1}(s)B_r^*(s)\Pi(s)]T_{K,r}(s, t)ds \end{aligned}$$

The following theorem, which generalizes a result of [12] to the time-varying situation, shows the existence of the optimal location within nuclear norm.

Theorem 3.1: For the time-varying system (12) with the uniformly bounded mild evolution operator $T(\cdot, \cdot)$ and the

cost functional (13), $B_r \in L_{s,\infty}(t_0, t_f; U, X)$, and for fixed t , $B_r(t)$, $r \in \Omega^m$ is a family of compact operators with the property that $\lim_{r \rightarrow r_0} \|B_r - B_{r_0}\|_{\infty} = 0$, for any $r_0 \in \Omega^m$, if U and Y are finite-dimensional, and G is nuclear, then the solution of corresponding integral Riccati equation $\Pi_r(t)$ are continuous of r in the nuclear norm

$$\lim_{r \rightarrow r_0} \|\Pi_r(t) - \Pi_{r_0}(t)\|_1 = 0, t \in [t_0, t_f],$$

and there exists an optimal location \hat{r} such that

$$\hat{\ell}_1 = \|\Pi_{\hat{r}}(t_0)\|_1 = \inf_{r \in \Omega^m} \|\Pi_r(t_0)\|_1$$

Similar results can be obtained for the discrete system. To be specified to CTM with emissions, we apply the approach to the model consisting of a combination of initial states and emissions.

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