

# Optimal reduction of anthropogenic emissions for air pollution control and the retrieval of emission source from observed pollutants III: Emission source inversion using a double correction iterative method

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**Abstract** Using the incomplete adjoint operator method in part I of this series of papers, the total emission source  $\mathbf{S}$  can be retrieved from the pollutant concentrations  $\mathbf{p}_{\text{ob}}$  obtained from the air pollution monitoring network. This paper studies the problem of retrieving anthropogenic emission sources from  $\mathbf{S}$ . Assuming that the natural source  $\mathbf{S}_n$  is known, and as the internal source  $\mathbf{S}_c$  due to chemical reactions is a function of pollutant concentrations, if the chemical reaction equations are complete and the parameters are accurate,  $\mathbf{S}_c$  can be calculated directly from  $\mathbf{p}_{\text{ob}}$ , and then  $\mathbf{S}_a$  can be obtained from  $\mathbf{S}$ . However, if the chemical reaction parameters (denoted as  $\gamma$ ) are insufficiently accurate, both  $\gamma$  and  $\mathbf{S}_c$  should be corrected. This article proposes a “double correction iterative method” to retrieve  $\mathbf{S}_c$  and correct  $\gamma$  and proves that this iterative method converges.

**Keywords** Air pollution, Anthropogenic emission source, Source due to chemical reaction, Source retrieval, Incomplete adjoint operator, Double correction iterative method

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## 1. Mathematical problem of retrieving the emission source using the known temporal and spatial pollutant concentrations

Ideally, the air pollution monitoring network can provide temporal and spatial concentration data for all air pollutants. This is not only necessary for air pollution monitoring, forecasting, air pollution control, and air quality management but can also be used to retrieve the temporal and spatial changes of emission sources and calibrate or correct the parameters in the air pollution chemical reaction equations. For this purpose, the advection-diffusion equations of air pollutants (the dynamic equations and the initial boundary

conditions) are as follows:

$$\begin{cases} \frac{\partial \mathbf{p}}{\partial t} = A(\mathbf{p}) + (\mathbf{S}_n + \mathbf{S}_a + \mathbf{S}_c(\mathbf{p})), \\ \Lambda(\mathbf{p})|_{\partial\Omega} = 0, \\ \mathbf{p}|_{t=0} = \mathbf{p}_{00}, \end{cases} \quad (1)$$

where  $\mathbf{p}$  is the  $n$ -dimensional vector for  $n$  pollutant concentrations,  $(\mathbf{S}_n, \mathbf{S}_a, \mathbf{S}_c)$  are the natural sources, anthropogenic emission sources, and sources due to atmospheric chemical reactions, respectively. The spatial area and its boundary are denoted as  $\Omega$  and  $\partial\Omega$ , respectively; and the time period is  $t \in [0, T]$ .  $A$  is the sum of the advection-diffusion and deposition operators, and  $\Lambda$  is the boundary condition operator.  $\mathbf{p}_{00}$  is the initial concentration values of the air pollutants in  $\Omega$ .

Despite the nonlinearity of the advection-diffusion equa-

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tions of air pollutants, in the case of heavy pollution, the solutions can be written as the sum of two parts: one part is the solution with a nonzero initial value condition ( $\rho_{00} \neq 0$ ), homogeneous boundary conditions, and linear  $\Lambda$ ; and the second part is the solution with nonhomogeneous boundary conditions and  $\rho_{00} = 0$  (Zeng and Wu, 2018). Eq. (1) is just the former with homogeneous boundary conditions. The 4-dimensional space constructed from area  $\Omega$  and time period  $t \in [0, T]$  is denoted as  $\Omega t$ , and the observed pollutant concentration  $\rho$  provided by the monitoring network is denoted as  $\rho_{ob}$ . The task of this paper is to retrieve the anthropogenic emission sources,  $S_a$ , using the known  $\rho_{ob}$ .

For the purpose of retrieving the corresponding  $S_a$  with the known  $\rho_{ob}$ , the objective function  $J(\rho, \rho_{ob})$  is defined as

$$J(\rho, \rho_{ob}) \equiv \int_0^T \iiint_{\Omega} \|\rho - \rho_{ob}\|^2 d\Omega dt, \quad (2)$$

then, the retrieval problem is finding the  $S_a$  that minimizes  $J(\rho, \rho_{ob})$ .

Referring to the method in Zeng and Wu (2018), the adjoint operator problem of retrieving  $S$  from  $\rho_{ob}$  is constructed as

$$\begin{cases} \frac{\partial \rho^*}{\partial t} = -A^*(\rho^*) - a\rho_{ob}, \\ \Lambda^*(\rho^*)|_{\partial\Omega} = 0, \\ \rho^*|_{t=T} = 0, \end{cases} \quad (3)$$

where  $A^*$  and  $\Lambda^*$  are the adjoint operators to  $A$  and  $\Lambda$  respectively; and  $a$  is required to balance the dimensions and units of both sides of eq. (3). Eq. (3) has the same form as the adjoint equations in Zeng and Wu (2018) and Zeng et al. (2020), but the difference lies in the nonhomogeneous term, which is  $a\rho_{ob}$  in this eq. (3), but  $aW$  in Zeng and Wu (2018) and Zeng et al. (2020). In addition, the values of  $a$  are also different.

$\rho^*$  can be obtained by solving eq. (3) with the known  $\rho_{ob}$ . Then, the  $S$  that satisfies eq. (2) can be obtained using the method in Zeng and Wu (2018). The next task is to retrieve  $S_a$  from  $\rho_{ob}$  and  $S$ .

## 2. Inversion of anthropogenic sources and correction of chemical reaction rate constants

Assuming that the natural source  $S_n$  is known, we define  $S' \equiv S - S_n$ . As  $S_c$  is a function of  $\rho$ , it can be obtained directly from  $\rho = \rho_{ob}$ ; thus,  $S_a$  can be obtained from the solved  $S$  in Section 1, and then,  $S_a = S' - S_c$ . However, the chemical reaction equations of  $S_c$  may be inaccurate. For example, the equations contain many chemical reaction rate constants  $\gamma$ , most of which are determined under laboratory conditions but may be inaccurate under actual environmental conditions. If  $\gamma$  is not corrected, the  $S_c$  calculated from  $\rho_{ob}$  will be

inaccurate and then affect the inversion accuracy of  $S_a$ . Therefore, we propose a method, called the “double correction iterative method” to correct  $\gamma$  and retrieve  $S_a$  simultaneously. The process is as follows.

Denoting the original value of  $\gamma$  as  $\gamma^{(0)}$ ,  $\rho_{ob}$  as  $\rho^{(0)}$ , and the  $S_c$  calculated from  $\gamma^{(0)}$  and  $\rho^{(0)}$  as  $S_c^{(0)}$ , then,  $S_a^{(0)} = S' - S_c^{(0)}$ . Substituting  $S_a^{(0)}$  into the air pollution advection-diffusion equations with the original chemical reaction equations, in which  $\gamma^{(0)}$  is used, we obtain the new values of  $\rho$  and  $S_c$ , denoted as  $\rho^{(1)}$  and  $S_{cc}^{(1)}$ , respectively.

If  $\|\rho^{(1)} - \rho_{ob}\|$  is very small, it is deduced that  $\|\rho^{(1)}\|$  is very close to  $\|\rho_{ob}\|$ , then  $\gamma^{(0)}$  is relatively accurate,  $S_{cc}^{(1)}$  is close to  $S_c^{(0)}$ ,  $S_a^{(0)}$  is close to  $S_a$ , and  $S_a = S_a^{(0)}$  is reliable. If  $\|\rho^{(1)} - \rho_{ob}\|$  and  $\|\rho^{(1)}\| - \|\rho_{ob}\|$  are beyond the allowable range,  $\gamma^{(0)}$ ,  $S_c^{(0)}$  and  $S_a^{(0)}$  are unreliable and needed to be corrected further.

Assuming that  $S_a$  does not contain the anthropogenic scavenging term, both the total value of the pollutant concentration  $\|\rho\|$  and the total value of the chemical reaction source  $\{S_c\}$  generally increases as the total value of the anthropogenic emission source  $\{S_a\}$  increases. This situation is described as Law A, which is the case discussed in this paper.  $\{S_a\}$  and  $\{S_c\}$  are defined as

$$\{S_a\} = \int_0^T \iiint_{\Omega} S_a d\Omega dt, \quad (4)$$

$$\{S_c\} = \int_0^T \iiint_{\Omega} S_c d\Omega dt. \quad (5)$$

$\|\rho^{(v)}\|$  is given as

$$\|\rho^{(v)}\| = (1 + \lambda^{(v)}) \|\rho_{ob}\|, \quad (6)$$

where  $v$  is iteration time, the correction of  $\gamma$  is as follows:

$$\gamma^{(v)} = (1 + \beta^{(v)} w) \gamma^{(0)}. \quad (7)$$

Then,

$$\|\gamma^{(v)}\| = \|\gamma^{(0)}\| + \beta^{(v)} \cdot (w, \gamma^{(0)}), \quad (8)$$

where  $\beta^{(v)}$  is the correction coefficient,  $w$  is the weight reflecting the importance of the reaction rate of each chemical reaction process (the contribution to the pollutant generation rate), and its components  $w \geq 0$ . The initial values of  $\lambda$  and  $\beta$  are taken as  $\lambda^{(0)} = 0$  and  $\beta^{(0)} = 0$ , respectively. In addition, when  $v = 0$ , eqs. (6) and (7) are self-consistent. When there are multiple pollutants, both  $\gamma$  and  $w$  are vectors.  $w$  can be determined by the chemical reaction equations and the sensitivity of the pollutant concentration to the chemical reaction process. The latter is similar to the method of sensitivity analysis of the air pollutant concentration to the emission sources (Liu et al., 2007; Liu and Huang, 2011). In practice, for less important chemical reaction processes,  $w$  can be ta-

ken as zero, and the most sensitive process has the largest  $w$ , which is denoted as  $w_M$ .

As  $S_c$  is the function of  $\gamma$  and  $\rho$ , denoted as  $S_c(\gamma, \rho)$ , we also have

$$S_c^{(v)} \equiv S_c(\gamma^{(v)}, \rho_{ob}), \quad (9)$$

$$S_a^{(v)} = S' - S_c^{(v)}. \quad (10)$$

In the iterative process, if there is one iteration to make  $\lambda^{(v)} < 0$ , that is,  $\|\rho^{(v)}\| < \|\rho_{ob}\|$ , then according to Law A,  $\{S_a^{(v-1)}\}$  is too small, and  $\{S_c^{(v-1)}\}$  is too large; thus,  $\gamma^{(v-1)}$  is too large, and  $\beta^{(v)}$  should be set as  $\beta^{(v)} < \beta^{(v-1)}$  to make  $\{S_a^{(v)}\} > \{S_a^{(v-1)}\}$ . However, if  $\lambda^{(v)} > 0$ , it is deduced that  $\{S_a^{(v-1)}\}$  is too large, and  $\{S_c^{(v-1)}\}$  is too small; thus,  $\gamma^{(v-1)}$  is too small, and  $\beta^{(v)}$  should be set as  $\beta^{(v)} > \beta^{(v-1)}$ .

Correspondingly,  $\beta^{(1)}$  can be taken as follows:

$$\begin{cases} -1/w_M < \beta^{(1)} < \beta^{(0)} = 0, & \text{if } \lambda^{(1)} < 0, \\ 0 = \beta^{(0)} < \beta^{(1)}, & \text{if } \lambda^{(1)} > 0. \end{cases} \quad (11)$$

Therefore, if  $\lambda^{(1)} < 0$ , starting from  $v=1$ ,  $\beta^{(v)}$  is set as  $\beta^{(v)} < \beta^{(v-1)}$  according to eq. (11). Then  $\gamma^{(v)}$  is obtained from eq. (7).  $\gamma^{(v)}$  and  $\rho_{ob}$  are substituted into eqs. (9) and (10) to obtain  $S_a^{(v)}$ , then  $\rho^{(v)}$  can be calculated and the sign of  $\lambda^{(v)}$  can be determined. Through this iteration, there is always one  $v$  ( $v \geq 2$ ) to make  $\lambda^{(v)} > 0$ . Then, from this  $v$ ,  $\beta^{(v+1)}$  should be set as  $\beta^{(v+1)} > \beta^{(v)}$ , i.e.,

$$\beta^{(v)} < \beta^{(v+1)} < \beta^{(v-1)} \quad (\text{when } \lambda^{(v-1)} < 0, \lambda^{(v)} > 0). \quad (12)$$

If  $\lambda^{(1)} < 0$ , starting from  $v=1$ ,  $\beta^{(v)}$  is set as  $\beta^{(v)} > \beta^{(v-1)}$ ; then  $\gamma^{(v)}$  is obtained from eq. (7).  $\gamma^{(v)}$  and  $\rho_{ob}$  are substituted into eqs. (9) and (10) to obtain  $S_a^{(v)}$ , then  $\rho^{(v)}$  can be calculated and the sign of  $\lambda^{(v)}$  can be determined. Through this iteration, there is always one  $v$  ( $v \geq 2$ ) to make  $\lambda^{(v)} < 0$ . Then, from this  $v$ ,  $\beta^{(v+1)}$  should be set as  $\beta^{(v+1)} < \beta^{(v)}$ , i.e.,

$$\beta^{(v-1)} < \beta^{(v+1)} < \beta^{(v)} \quad (\text{when } \lambda^{(v-1)} > 0, \lambda^{(v)} < 0). \quad (13)$$

Continuing this iteration, the length of the interval ( $\beta^{(v)}$ ,  $\beta^{(v+1)}$ ) and the value of  $|\lambda^{(v)}|$  will decrease until  $\|\rho^{(v \rightarrow \infty)}\| - \|\rho_{ob}\| < \varepsilon$ , where  $\varepsilon$  is any given small positive number. Therefore,  $\lambda^{(v)}$  is a convergent sequence and  $S_a^{(v)}$  is

the retrieved  $S_a$  as  $v \rightarrow \infty$ , and the correction of the chemical reaction rate constants is also given as  $\gamma^{(\infty)} = (1 + \beta^{(\infty)} w) \gamma^{(0)}$ .

### 3. Conclusions and discussions

In Section 2, we retrieved the emission source, which can provide a better time-varying emission source distribution; and corrected some parameters in the chemical reaction equations. If the concentration of each pollutant can be obtained comprehensively and the monitoring network is sufficiently dense and accurate, this method works well for both source inversion and parameter corrections. However, the correction method here is similar to the parameter adjustment method in numerical simulation and is disadvantageous to only find out some applicable parameters. To truly solve the chemical reaction problem and obtain reliable parameters for the actual environment, the kinetics of the chemical reaction process in different situations should be studied. Additionally, if it is difficult to obtain sufficiently dense, complete and accurate data of  $\rho_{ob}$ , some monitoring data of  $S_a$  are additionally required for the inversion; and data assimilation methods should be employed, such as the work in Zhu and Wang (2006).

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