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Optimal reduction of anthropogenic emissions for air pollution control and the retrieval of emission source from observed pollutants I. Application of incomplete adjoint operator

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Abstract The ultimate solution to anthropogenic air pollution depends on an adjustment and upgrade of industrial and energy structures. Before this process can be completed, reducing the anthropogenic pollutant emissions is an effective measure. This is a problem belonging to "Natural Cybernetics", i.e., the problem of air pollution control should be solved together with the weather prediction; however, this is very complicated. Considering that heavy air pollution usually occurs in stable weather conditions and that the feedbacks between air pollutants and meteorological changes are insufficient, we propose a simplified natural cybernetics method. Here, an off-line air pollution evolution equation is first solved with data from a given anthropogenic emission inventory under the predicted weather conditions, and then, a related "incomplete adjoint problem" is solved to obtain the optimal reduction of anthropogenic emissions. Usually, such solution is sufficient for satisfying the air quality and economical/social requirements. However, a better solution can be obtained by iteration after updating the emission inventory with the reduced anthropogenic emissions. Then, this paper discusses the retrieval of the pollutant emission source with a known spatio-temporal distribution of the pollutant concentrations, and a feasible mathematical method to achieve this is proposed. The retrieval of emission source would also help control air pollution.

Keywords Air pollution, Optimal control, Source retrieval, Incomplete adjoint operator

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1. Introduction

The solution to anthropogenic air pollution ultimately depends on an adjustment and upgrade of the industrial and energy structures. Before this process can be completed, reducing the pollution emissions is the effective measure. Therefore, government should establish feasible emission reduction strategies that can meet the air quality standards, meanwhile minimizing the economic loss due to the emission reduction, or maximizing the social welfare. This optimal air quality management can be referred to as "optimal control", and can be achieved by the natural cybernetics method (Zeng, 1996).

Anthropogenic air pollution is obviously affected by atmospheric and environmental processes such as weather processes. Therefore, weather processes must be considered when regulating air pollution conditions. In other words, emission reduction strategies must be established with weather forecasts. This is the essential difference between the methods of regulating natural or environmental processes (one subject of natural cybernetics) and engineering cybernetics (Zeng, 1996; Zhu et al., 1999; Zhu and Zeng, 2003; Zeng et al., 2012; Huang et al., 2014). However, the natural cybernetics approach needs massive computing. Although the control of air pollution by natural cybernetics has for-

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merly been proposed (Zeng, 1996; Zhu and Zeng, 2003), it can now be achieved with the new-age highly developed supercomputing. This paper presents an optimal control method that is more realizable than formerly proposed methods.

The anthropogenic emissions of air pollutants (i.e., anthropogenic sources) are actually dynamic, as they depend on time and location, and it is difficult to accurately assess its spatio-temporal distribution. Generally, an average inventory of gridded anthropogenic emissions throughout a certain period has been estimated based on the information and emission factors in different sectors (Zhang et al., 2006), and this approach is referred to as the "bottom-up" approach. The "bottom-up" emission inventory is updated slowly and has some omissions and uncertainties. Fortunately, because of new developments in air quality monitoring networks, the spatio-temporal distributions of pollutant concentrations can be assessed more accurately. Therefore, a "top-down" inversion method can be developed to estimate the source emissions, which is important for pollution control. In the second part of this paper (Section 4), we study this inversion method and provide a mathematical solution.

2. The mathematical problem to predict air pollution

After the pollutants are emitted into the atmosphere, their distribution and evolution depend on atmospheric motions, e.g. advection and diffusion, as well as many physical and chemical processes, which can generate new types of pollutants. In principle, these have been described by the complicated Atmospheric Chemistry and Transport Model (ACTM). And the air pollution can be predicted using ACTM coupled with Numerical Weather Prediction (NWP). The ACTM has been developed as both the "on-line" way that is coupled with NWP, and as "off-line" way, where the NWP results are used as the ACTM input meteorology data. Both models have been used in routine operations (Byun and Schere, 2006; Luo and Wang, 2006; ENVIRON, 2012). The optimal control based on natural cybernetics should be the same. However, the ACTM coupled with NWP is complicated and consists of very complex nonlinear partial differential equations, which makes it very time-consuming. As the heavy air pollution generally occurs under the stable weather conditions; thus, the first approximation of an off-line model could be used for the optimal control of air pollution where the solution is approximated by using ACTM alone but with known meteorological conditions, such as the air velocity $\vec{V} = u\vec{i} + v\vec{j} + w\vec{k}$ and turbulent mixing parameters $\vec{K} = k_x\vec{i} + k_y\vec{j} + k_z\vec{k}$ (usually $k_x = k_y$), which can be provided by NWP. In this off-line model, the air pollutant concentration ρ is described by the following equations and initial and boundary conditions:

$$\frac{\partial \rho}{\partial t} = L(\rho) + D(\rho) + S, \tag{1}$$

$$\begin{aligned} L(\rho) &= -V \cdot \nabla \rho + \left(\frac{\partial}{\partial x} k_x \frac{\partial \rho}{\partial x} + \frac{\partial}{\partial y} k_y \frac{\partial \rho}{\partial y} + \frac{\partial}{\partial z} k_z \frac{\partial \rho}{\partial z} \right) \\ &\equiv -V \cdot \nabla \rho + \nabla \cdot (K \cdot \nabla \rho), \end{aligned} \tag{2}$$

where $\rho = (\rho_1, \rho_2, \cdots, \rho_n)$ is a vector with n pollutant species $\rho_i (i = 1, 2, \cdots, n)$ as its components, L is the advection plus turbulent mixing, D is the deposition, and $S = S_n + S_a + S_c$, S_n and S_a are the natural emissions and anthropogenic emissions, respectively, S_c is the internal source that emerged from chemical reactions. Most components of S_c are complicated nonlinear operators depending on ρ, while L and D are linear operators. The inputs S_n, S_a, and S_c are difficult to be separated.

The air pollutant concentration ρ is defined over an area Ω, and the boundaries of Ω are shown in Figure 1, where $\partial_1\Omega$ is the bottom boundary with $z = z_s$ (z_s is the height above ground level), and $\partial_2\Omega$ is the lateral boundary. Therefore, the boundary conditions are given as

$$\begin{cases} \rho|_{t=0} = \rho_0(x, y, z) \\ \rho|_{\partial_2 \Omega} = \rho_b|_{\partial_2 \Omega} \\ k_z(\partial \rho / \partial z)|_{\partial_1 \Omega} \to 0 \\ k_z(\partial \rho / \partial z)|_{z\to\infty} \to 0,\ w\rho|_{z\to\infty} \to 0 \end{cases}, \tag{3}$$

where ρ_b means the background pollution.

There is no lateral boundary $\partial_2\Omega$ if Ω covers the whole global region, but for regional air quality forecasting and control, $\partial_2\Omega$ can be set at a region far away from the anthropogenic pollution where the background pollution is small. For example, for a heavy pollution case in the northern China (Beijing-Tianjin-Hebei area), Figure 2 shows that $\partial_2\Omega$ can be set at the Taihang Mountains and along the Yellow sea southwest into Henan Province, while the other two $\partial_2\Omega$ can be set at the borders of Northeast China and the Shanxi to Henan Province.

It can be proven that, at the given S, the solution ρ of Eqs. (1)–(3) exists and is unique, and $\rho \geq 0$ in Ω as $S \geq 0$ and is denoted later as ρ_0.

3. The mathematical problem of optimal reduction of anthropogenic emission

The emission reduction requires that the air quality reaches a given level in the domain $\Omega' \subseteq \Omega$. First, we try to reduce the S. Suppose the current emission is S_a and the reduction is S' ($S' \geq 0$). Therefore, $\rho = \rho_0 - \rho'$, and ρ' satisfies the following equations:

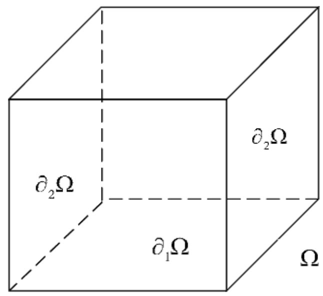

Figure 1 The solution area and the boundaries.

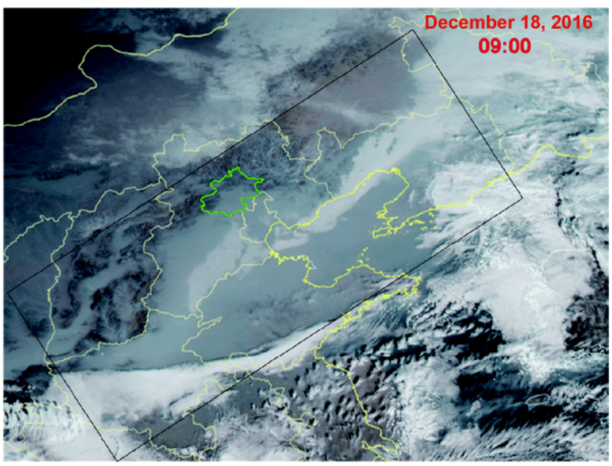

Figure 2 The satellite view of a heavy pollution case in Beijing-Tianjin-Hebei area. The light gray area over the North China is covered by a heavy haze, and the white area is covered by a cloud or fog. The rectangle shows the suggested lateral boundary of $\partial\Omega$.

$$\frac{\partial \rho'}{\partial t} = L(\rho') + D(\rho') + S', \tag{4.1}$$

$$\Lambda_2(\rho') = \rho'|_{\partial_2\Omega} = 0, \tag{4.2}$$

$$\Lambda(\rho') = \left.\frac{\partial \rho'}{\partial z}\right|_{\partial_1\Omega} = 0, \quad \left.\frac{\partial \rho'}{\partial z}\right|_{z\to\infty} \to 0, \quad w\rho'|_{z\to\infty} \to \rho, \tag{4.3}$$

$$\rho'|_{t=0} = 0. \tag{4.4}$$

$\rho|_{t=0} = \rho_0$ and $\rho|_{\partial_2\Omega} = \rho_b$; therefore, $\rho'|_{t=0} = \rho'|_{\partial_2\Omega} = 0$. It can be proved that the solution ρ' exists and $\rho' \geq 0$ when $S' \geq 0$; then S' can be deduced under the specific economic loss constraints.

3.1 Mathematical formulations of objective and constraint

The objective of the emission reduction is to ensure the air quality reaches the satisfied level in the domain $\Omega' \subseteq \Omega$. Here, we require:

$$\int_0^T \iiint_{\Omega'} W\rho \, \mathrm{d}\Omega \mathrm{d}t = \int_0^T \iiint_{\Omega'} W\left(\rho_0 - \rho'\right) \mathrm{d}\Omega \mathrm{d}t \leq D, \tag{5}$$

which leads to

$$(W, \rho') \equiv \int_0^T \iiint_{\Omega''} W\rho' \, \mathrm{d}\Omega \mathrm{d}t \geq \int_0^T \iiint_{\Omega''} W\rho_0 \mathrm{d}\Omega \mathrm{d}t - D \equiv D' \geq 0, \tag{6.1}$$

where $\Omega'' \subseteq \Omega$ is the domain where the emission reduction is required, and $W(t;\, x,\, y,\, z)$ is the weight, which is chosen according to the importance of the areas. $W = 1$ when each area is addressed equally.

Besides, the economic loss caused by emission reduction is required to be minimized; that is,

$$\left(E_C, S'\right) \equiv \int_0^T \iiint_{\Omega''} E_C S' \, \mathrm{d}\Omega \mathrm{d}t = \min, \tag{6.2}$$

where $E_C(t;\, x,\, y,\, z) \geq 0$ is the unit economic loss caused by the emission reduction, which can be determined through the industry's economics. Note, $E_C = 1$ when the loss of the different emission reduction is the same. Below, we take Ω' and Ω'' as the Ω by intruding W and E_C, where $W = 0$ outside Ω' and $E_C = 0$ outside Ω''.

The formulation consisting of Eqs. (6.1) and (6.2) is one of the objective options applicable for air pollution control. When Eq. (6.1) is not available, the other objective formulation should be used to generally maximize the health and social benefits.

For a given S', Eq. (4) is the linear problem. An incomplete adjoint operator problem of Eq. (4) is

$$\frac{\partial (E_C\rho^*)}{\partial t} = -\Lambda^*\left(E_C\rho^*\right) - \mathrm{a}W, \tag{7.1}$$

$$\Lambda^*\left(E_C\rho^*\right) = 0, \quad \text{on the } \partial\Omega, \tag{7.2}$$

$$E_C\,\rho^*\big|_{t=T} = 0, \tag{7.3}$$

where $A^* = L^*\left(E_C\,\rho^*\right) + D^* = \nabla\cdot\left(E_C\,\rho^* V\right) + \nabla\cdot(K\cdot\nabla[E_C\,\rho^*]) + D^*$, $D^*$is the adjoint operator to D and can be formulated out when D is given, $\Lambda^* = \Lambda$, and constant a is required to balance the dimensions and units. Here "incomplete" means that Eq. (7) is the adjoint to Eq. (4) with a given S', but S' nonlinearly depends on ρ'. The key idea to solve Eq. (7) is directly connecting $(W,\, \rho')$ with $(E_C,\, S')$. Given E_C and W, ρ^* can be solved by the adjoint equation and is unique, but S' remains unknown.

3.2 Determining the optimal emission reduction strategy

Multiplying both sides of Eqs. (4.1) and (7.1) by $E_C\rho^*$ and ρ', respectively, and integrating over (0–T) × Ω, we have

$$0=\int_0^T\iiint_\Omega [E_C\rho^*A(\rho')-\rho' A^*(E_C\rho^*)]\mathrm{d}\Omega\mathrm{d}t+\int_0^T\iiint_\Omega (E_C\rho^*S'-\mathrm{a}\rho' W)\mathrm{d}\Omega\mathrm{d}t. \quad (8)$$

The first integration is 0 by the boundary conditions in Eqs. (4.2), (4.3), and (7.2), and the second integration is also 0. So we have

$$\int_0^T\iiint_\Omega (\mathrm{a}W\rho')\mathrm{d}\Omega\mathrm{d}t=\int_0^T\iiint_\Omega \left(E_C S'\rho^*\right)\mathrm{d}\Omega\mathrm{d}t. \quad (9)$$

Therefore, the integrations of a$W\rho'$ and $E_C S'\rho^*$ are connected directly. According to Eqs. (6.1) and (9), we have

$$\int_0^T\iiint_\Omega \mathrm{a}W\rho'\mathrm{d}\Omega\mathrm{d}t=\int_0^T\iiint_\Omega (E_C\rho^*)S'\mathrm{d}\Omega\mathrm{d}t\geq \mathrm{a}D'>0, \quad (10)$$

i.e., the inner product $(E_C S', \rho^*) \geq \mathrm{a}D'$. Let the projection of $E_C S'$ onto ρ^* be $\gamma\rho^*$ (where γ is a coefficient), the left part $E_C S'_\perp$ is orthogonal to ρ^*, and $\left(E_C S'_\perp,\rho^*\right)=0$. Therefore, $S'_\perp$ has negative values, and it is hard to ensure $\gamma\rho^*+E_C S'_\perp$ is always positive. It is better to take$S'_\perp\equiv 0$, so that we have

$$S'=\gamma\frac{\rho^*}{E_C}. \quad (11)$$

Introducing $\|\rho^*\|$ and M^* as follows,

$$\|\rho^*\|^2\equiv\int_0^T\iiint_\Omega (\rho^*)^2\mathrm{d}\Omega\mathrm{d}t, \quad (12.1)$$

and

$$M^*\equiv\int_0^T\iiint_\Omega \rho^*\mathrm{d}\Omega\mathrm{d}t. \quad (12.2)$$

Therefore, the requirements of Eqs. (6.1) and (6.2) respectively become

$$\gamma\|\rho^*\|^2\geq \mathrm{a}D', \quad (13.1)$$

and

$$\gamma M^*=\min. \quad (13.2)$$

Hence, we have

$$\gamma=\frac{\mathrm{a}D'}{\|\rho^*\|^2}. \quad (14)$$

Substituting γ into Eq. (11), the optimal emission reduction S' is obtained. The next step is to obtain S'_a, the optimal reduction of anthropogenic emission, from the equation $S'=S'_a+S'_c$ by iteration. This will be described in next paper (Ⅱ). It is also possible to obtain a nontrivial solution $S_\perp$, but the procedure is rather complicated (Zeng, 1995).

4. Source retrieval from the observed air pollutant data

Let the spatio-temporal domain Ωt be defined by $t\in[0,T]$ and $(x, y, z)\in\Omega$. Supposing that the air pollutant ρ in Ωt is given by the observational data, the problem is how to retrieve the source S. Denote S as S' in the followings. Given weather conditions in Ωt, let the solution of the Eqs. (1)–(3) with S = 0 be ρ_0, and the solution of the same problem with trivial initial and boundary conditions and S = S' be ρ'; i.e., $\rho'|_{t=0}=0$, $A(\rho')=0$, but $S=S'$. We denote the latter problem as Eq. (4)′, so that S' is solved.

Let the observed ρ be $\rho_{\mathrm{ob}}(x,y,z,t)$. Denote $\rho_{\mathrm{ob}}-\rho_0\equiv\rho'_{\mathrm{ob}}$; set up the incomplete adjoint problem analogous to Eq. (7) but with E_C = 1, and replace aW with $\mathrm{a}\rho'_{\mathrm{ob}}$. Thus, we have

$$\begin{cases}\dfrac{\partial\rho^*}{\partial t}=-A^*(\rho^*)-\mathrm{a}\rho'_{\mathrm{ob}},\\ A^*\rho^*=A\rho^*=0,\\ \rho^*|_{t=T}=0,\end{cases} \quad (15)$$

where the constant a is also introduced to balance the dimensions and units of both sides.

We require that $\rho_0+\rho'$ is mostly closed to ρ_{ob}, leading to

$$\left\|\rho'_{\mathrm{ob}}-\rho'\right\|^2_{\Omega t=(0\sim T)\times\Omega}\equiv\int_0^T\iiint_\Omega \left|\rho'_{\mathrm{ob}}-\rho'\right|^2\mathrm{d}\Omega\mathrm{d}t=\min, \quad (16)$$

and

$$\|\rho'\|^2_{\Omega t}=\left\|\rho'_{\mathrm{ob}}\right\|^2_{\Omega t}. \quad (17)$$

Hence, we have

$$\|\rho'\|^2+\left\|\rho'_{\mathrm{ob}}\right\|^2-2\left(\rho',\rho'_{\mathrm{ob}}\right)=2\left[\left\|\rho'_{\mathrm{ob}}\right\|^2-\left(\rho',\rho'_{\mathrm{ob}}\right)\right]=\min. \quad (18)$$

This means the two requirements of Eqs. (16) and (17) lead to

$$\left(\rho',\rho'_{\mathrm{ob}}\right)=\max. \quad (17)'$$

Eq. (4)′×ρ^* and Eq. (16)×ρ' respectively yield

$$\rho^*\frac{\partial\rho'}{\partial t}-\rho^*A(\rho')=\rho^*S', \quad (19)$$

and

$$\rho'\frac{\partial\rho^*}{\partial t}+\rho'A^*(\rho^*)=-\mathrm{a}\rho'\rho'_{\mathrm{ob}}. \quad (20)$$

Integrating Eqs. (19) and (20) over Ωt and summing together, we yield

$$\int_0^T\iiint_\Omega \rho^*S'\mathrm{d}\Omega\mathrm{d}t=\int_0^T\iiint_\Omega \mathrm{a}\rho'\rho'_{\mathrm{ob}}\mathrm{d}\Omega\mathrm{d}t. \quad (21)$$

Therefore, Eq. (17)′ is transformed into

$$\int_0^T\iiint_\Omega \rho^*S'\mathrm{d}\Omega\mathrm{d}t=\max. \quad (22)$$

Let $S'_{//}$ and $S'_{\perp}$ are the parallel and orthogonal projections to ρ^*, respectively; therefore, we have $S'_{//} = \gamma\rho^*$, and

$$S' = \gamma\rho^* + S'_{\perp}, \tag{23}$$

where γ is a coefficient to be determined, while $\left(S'_{\perp}, \rho^*\right) = 0$.

Let $\rho'_{//}$ and $\rho'_{\perp}$ be the two solutions to Eq. (4) as S' is equal to $S'_{//}$ and $S'_{\perp}$, respectively. Substituting $\rho' = \rho'_{//} + \rho'_{\perp}$ into Eq. (21) yields $(\rho^*, S'_{//}) = (\rho'_{//}, \rho'_{\rm ob})$, and

$$0 = \left(\rho^*, S'_{\perp}\right) = \left(\rho'_{\perp}, \rho'_{\rm ob}\right). \tag{24}$$

This means that $\rho'_{\perp}$ is orthogonal to $\rho'_{\rm ob}$ and represents some insignificant amount of noise. So we can take $S' = S'_{//}$ and $\rho' = \rho'_{//}$, and from Eq. (24), we obtain

$$\rho' = \rho'_{//} = \beta\rho'_{\rm ob}. \tag{25}$$

Substituting Eq. (25) into Eq. (17) yields $\beta = 1$, and substituting Eqs. (25) and (23) to both sides of Eq. (21) yields

$$\mathrm{a}\left\|\rho'_{\rm ob}\right\|^2 = \gamma\|\rho^*\|^2. \tag{26}$$

So, $\gamma = \mathrm{a}\left\|\rho'_{\rm ob}\right\|^2 / \|\rho^*\|^2$, and finally we obtain

$$S' = S'_{//} = \mathrm{a}\frac{\left\|\rho'_{\rm ob}\right\|^2}{\|\rho^*\|^2}\rho^*. \tag{27}$$

Considering that $\rho^* \geq 0$, $S'_{//}$ is the S' parallel projection onto ρ^*, and $\gamma > 0$, and $S'_{\perp}$ is orthogonal to ρ^*, there is some domain with $S'_{\perp} < 0$, and it is difficult to guarantee $S' = S'_{//} + S'_{\perp}$ is positive everywhere. These analyses and discussions lead to the conclusion that $S' = S'_{//}$ is the proper solution that satisfies the requirements of Eqs. (16) and (17). The method to obtain S'_a from S' will be described in detail in the next paper (Ⅱ).

5. Discussions and conclusions

Natural cybernetics is proposed to regulate and control subject (phenomenon or process) occurring in the nature, which is generally unsteady. Therefore, natural cybernetics involves predicting the evolution of the natural environment and studying its multi-interactions by coupled model; this makes it essentially different from engineering cybernetics. However, as natural cybernetics is applied to very practical problems that require time effectiveness, the solution should be simple and efficient. So, we propose a simplified solution by an incomplete adjoint operator in the first part of this paper, where we adopt an off-line model that does not consider the interaction between air pollutants and meteorological evolution. However, the emission reduction strategy obtained by this simplified method needs to be validated. This can be done by updating the emission inventories with the reduced emissions, and the ACTM coupled with NWP is used to get new pollutant concentrations and distributions. If the results meet the air quality standard in Eq. (5), this emission reduction strategy can be applied (perhaps it is not optimal, but it is good enough and applicable); otherwise, the method is iterated until the requirement of Eq. (5) is satisfied.

In the source retrieval problem, as the meteorological conditions are known, the off-line ACTM can be used with the adjoint operator to obtain the emission source from the air pollutant monitoring data. There are also other approaches to infer the dynamic emission source, for example, those given by Liu and Huang (2011) and their follow-up study. If the meteorological condition is unknown, the retrieval will be much more difficult. However, if meteorological data, pollutant concentration data, and emission source data are available for regions $(\Omega t)_1$, $(\Omega t)_2$, and $(\Omega t)_3$, respectively, they can be interpolated into a whole region Ωt using data assimilation methods. The accuracy of the result depends on the quantity and quality of the available data. A similar problem has been thoroughly investigated by Zeng (1995) and Zhu and Wang (2006).

It is necessary to note that Eq. (1) is a nonlinear partial differential equation as the chemical processes S_c included in S are nonlinear, so that in general, the solution ρ is different from $\rho_0+\rho'$, where ρ_0 and ρ' are the solutions to the problem with initial and boundary conditions, but without emission source S and with emission source ($S \not\equiv 0$), respectively. However, our simulation experiments show that for a heavy pollution, the difference is very small, and the nonlinearity appears weak (this will be published in another article), so the proposed methods with the adjoint operator for optimal emission reduction and dynamic emission source estimation are applicable in practice.

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