

FLUCTUATIONS IN TRACE GAS CONCENTRATIONS GENERATED BY ENTRAINMENT IN THE BOUNDARY LAYER AND FREE TROPOSPHERE*

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1. Introduction

Many trace gases in the atmosphere are emitted into the planetary boundary (PBL) near or at the surface and destroyed by chemical reactions as they move about in the atmosphere. These processes generate fluctuations in concentration which can be used to infer residence times of species, or alternatively, if the residence times are known, we may be able to infer transport properties of the atmosphere, or surface emission patterns. One way to study this is to analyze the variations of a suite of chemical species sampled at various locations and at various times. For example, an airplane can be used to collect samples during a flight, or series of flights, and the standard deviations of the species concentrations σ_i obtained from the collection of samples, normalized by their mean concentrations S_i , can be calculated. An application of this is to estimate residence times of trace species which are unknown by using a suite of species of different residence times τ_i to establish a relationship between σ_i/S_i and τ .

This procedure was apparently first described by Junge (1963 and 1974) who suggested that σ_i/S_i should be inversely proportional to τ_i ; i.e.,

$$\sigma_i/S_i = A\tau_i^{-\alpha} \quad (1)$$

with A a constant and $\alpha = 1$. Definitive tests of this hypothesis are only now being reported. Jobson, et al. (1999) have analyzed several archived data sets of quite extensive observations collected under different conditions and found that for measurements in the remote troposphere (i.e. far from localized sources), the value of α was about 1/2. Colman et al. (1999) used data from the NASA PEM-Tropics-A (Pacific Exploratory Mission) aircraft flights in 1996 to estimate the lifetime of

CH_3Br , and obtained a value of α considerably less than one. Ehhalt et al. (1998) used data from PEM West B to infer the concentration of OH from the residence times of a suite of hydrocarbons which react with OH and found a value of $\alpha = 0.48$, very close to 1/2.

Ehhalt et al. (1998) also developed a hierarchy of scalar diffusion models to illustrate aspects of diffusion of reactive species. They start with a simple one-dimensional model of steady-state vertical diffusion with a constant turbulent diffusivity that illustrates the inherent property of a second-order diffusion equation to give a mean concentration that is proportional to $\tau^{1/2}$. They then showed, using 2- and 3-dimensional models, that for continental sources of species they could simulate fluctuations in species concentrations that followed (1) with $\alpha \simeq 0.5$.

Here I develop a model, also using the concept of an eddy diffusion equation, but adding to this the process of mass exchange across the top of the PBL, as well across the tropopause, to study in more detail the behavior of trace reactive species in the PBL, as well as the overlying free atmosphere. This global model assumes horizontal homogeneity, so the only source of fluctuations is vertical exchange.

2. Model

The basis for this model is that mass exchange across the top of the PBL, as well as across the tropopause, can generate fluctuations in species concentrations in the PBL and free atmosphere. We parameterize this by constructing a model which consists of three layers: a lower layer (L1) that is well-mixed by direct interactions with the surface, assumed to be 1 km deep (the PBL); a middle layer (L2), which is the rest of the troposphere extending up to 15 km, where vertical transport is described by a constant diffusivity; and an overlying layer (L3), the stratosphere, assumed to extend up to infinity and described by a diffusivity which increases with height. Each of these layers is assumed to be separated by a dis-

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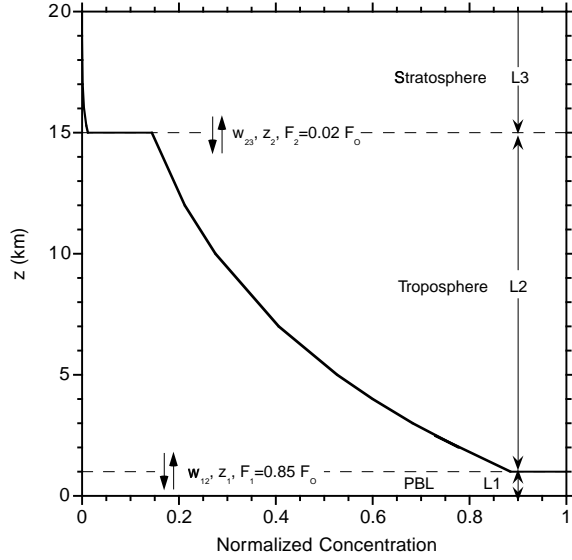


Figure 1: Modeled vertical profile from the surface up into the stratosphere of a reactive species released in the PBL with a residence time $\tau = 10^7$ s \simeq 116 days normalized by the PBL concentration.

continuous change in concentration across an interface where the transport is assumed to occur via an exchange velocity \times a change in concentration across the interface. Thus, the vertical flux across the top of L1 is

$$F_1 = w_{12}(S_1 - S_2). \quad (2)$$

Figure 1 shows the locations and names of the layers and interfaces.

The one-dimensional conservation equation for a species S with an atmospheric residence time τ is

$$\frac{\partial S}{\partial t} + \frac{1}{\rho} \frac{\partial \rho F}{\partial z} - \frac{S}{\tau} = 0, \quad (3)$$

where ρ is air density. We assume steady state. For L1 we assume a perfectly mixed PBL and $\tau \gg z_1/w_*$, where w_* is the convective velocity scale. That is, the residence time is assumed to be \gg the mixing time scale within the PBL, which is typically less than an hour. Then integrating (3) from the surface to the top of the PBL z_1 we obtain

$$F_1 - F_0 = \frac{z_1}{\tau} S_1. \quad (4)$$

For L2, we assume a constant eddy diffusivity $K_2 = 10$ m² s⁻¹ (Massie and Hunten, 1981). Substituting $F_2(z) = -K_2 S_2'(z)$ into (3), we obtain

$$\frac{1}{\rho} \frac{\partial}{\partial z} \rho K_2 \frac{\partial S_2(z)}{\partial z} - \frac{1}{\tau} S_2(z) = 0. \quad (5)$$

We use the approximation that $\rho \simeq \rho_0 e^{-\ell_2 z}$, where $\ell_2 \simeq 0.1134 \times 10^{-3}$ m⁻¹ for a standard troposphere, so that (5) becomes

$$\frac{\partial^2 S_2(z)}{\partial z^2} - \ell_2 \frac{\partial S_2(z)}{\partial z} - \frac{1}{K_2 \tau} S_2(z) = 0. \quad (6)$$

For L3, we assume $K_3(z) = K_3(z_2) e^{kz}$, where $K_3(z_2) = 0.0711$ m² s⁻¹ and $k = 0.103 \times 10^{-3}$ m⁻¹ (Liley, 1995). For density, we assume a standard isothermal stratosphere, $\ell_3 \simeq 0.157 \times 10^{-3}$ m⁻¹. Then,

$$\frac{\partial^2 S_3(z)}{\partial z^2} + (k - \ell_3) \frac{\partial S_3(z)}{\partial z} - \frac{1}{K_3(z_2) \tau} e^{-kz} S_3(z) = 0. \quad (7)$$

At the top of the PBL, I estimate the entrainment velocity (2) from the Earth's surface energy budget (neglecting shear). Sellers (1965) estimated the average sensible energy flux over land and ocean as 31.9 W m⁻² and 10.6 W m⁻², respectively, and the corresponding latent energy fluxes as 33.2 and 98.3 W m⁻², respectively, with 70.8% of the Earth covered by ocean. This leads to an average surface virtual temperature flux of 0.023 m s⁻¹ K. Applying the simple Tennekes (1973) mixed-layer model to estimate the jump in temperature across the PBL top,

$$T_2 - T_1 \simeq \frac{\gamma z_1}{2 + m^{-1}}, \quad (8)$$

where $-m$ is the ratio of virtual temperature flux at z_1 to the surface flux and γ is the lapse rate of potential temperature just above z_1 . Here I assume $m = 0.2$ and $\gamma = 6.5$ K km⁻¹. From (2), (8), and the surface virtual temperature flux, we estimate $w_{12} \simeq 0.005$ m s⁻¹. This gives a tropospheric turnover time of about 20 days.

At the top of the troposphere, we estimate the entrainment velocity w_{23} from the estimate of Holton et al. (1995) of a two-year turnover time of air above the 100 hPa level. This leads to an exchange velocity of $w_{23} \simeq 10^{-4}$ m s⁻¹.

Additional boundary conditions are provided by flux continuity across the interface between L1 and L2,

$$F_1(z_1) = w_{12}(S_1 - S_2(z_1)) = -K_2 S_2'(z_1), \quad (9)$$

and between L2 and L3,

$$\begin{aligned} F_2(z_2) &= w_{23}(S_2(z_2) - S_3(z_2)) = -K_2 S_2'(z_2) \\ &= -K_3(z_2) S_3'(z_2). \end{aligned} \quad (10)$$

A solution to (6) is

$$\begin{aligned} S_2(z) &= C_1 \exp \left[\left(\ell_2 - \sqrt{\ell_2^2 + \frac{4}{K_1 \tau}} \right) \frac{z}{2} \right] \\ &+ C_2 \exp \left[\left(\ell_2 + \sqrt{\ell_2^2 + \frac{4}{K_1 \tau}} \right) \frac{z}{2} \right] \end{aligned} \quad (11)$$

where C_1 and C_2 are constants to be determined by the boundary conditions. A solution to (7) is

$$S_3(z) = e^{\frac{\ell_3 z}{2}} [C_3 I_{-\nu}(\zeta) + C_4 I_{\nu}(\zeta)] \quad (12)$$

where $\nu = (\ell_3 - k)/k$,

$$\zeta = -\frac{2}{k} \sqrt{\frac{e^{-kz}}{K_3(z_2)\tau}}, \quad (13)$$

$I_{\pm\nu}(\zeta)$ are modified Bessel functions of order ν , and C_3 and C_4 are constants.

Scalar fluctuations in a horizontally homogeneous PBL are generated by fluxes at the surface and top of the PBL. This concept of bottom-up and top-down scalar diffusion in the convective PBL was developed by Wyngaard and Brost (1984). For the case of a reactive species released into the PBL at a location remote from the observation point with no surface deposition, PBL fluctuations result only from exchange of air with different values of S across the PBL top. The top-down formulation obtained from large-eddy simulation of the convective PBL by Moeng and Wyngaard (1989) predicts that

$$\sigma = \frac{F_1(z_1)}{w_*} f_t^{1/2}(z/z_1) \quad (14)$$

where $f_t(z/z_1)$ is the top-down variance function,

$$f_t(z/z_1) = 3.1(1 - z/z_1)^{-3/2}. \quad (15)$$

Substituting (9) into (14) yields

$$\sigma = \frac{w_{12}[S_1 - S_2(z_1)]}{w_*} f_t^{1/2}(z/z_1). \quad (16)$$

We see that as $z \rightarrow z_1$ or $w_* \rightarrow 0$, $f_t(z/z_1)$ becomes large. That is, as we approach z_1 , or as the turbulence becomes small, the variance becomes large. However, it cannot realistically exceed $[(S_1 - S_2(z_1))]$ unless significant gradients in S exist above or below z_1 .

Here we consider that σ both above and below z_1 results not only from small-scale turbulent mixing, as parameterized in (14), but also mesoscale circulations that exchange air between L1 and L2. Such exchange cannot mix out as rapidly as small-scale exchange, so we expect that σ is more directly related to the concentration difference between the two layers. Furthermore, we expect that larger fluctuations may exist in L2 than L1 since mixing is less and S can vary with height in L2. Therefore, we parameterize the standard deviation by

$$\sigma = C [S_1 - S_2(z_2)] \quad (17)$$

where C is a constant ≤ 1 in the PBL.

3. Results

Equations (4), (9), (10), (11), and (12) were solved for $\tau = 10^7$ s $\simeq 116$ days and the results for the mean concentrations are plotted in Figure 1. This value of τ was selected because it produces concentration fluctuations in the PBL and troposphere, with relatively low stratospheric and upper tropospheric concentration. We can approximate (11) by

$$S_2(z) \simeq S_2(z_2) e^{-z/\sqrt{K_2\tau}}, \quad (18)$$

and thus

$$S_2'(z) = -\frac{S_2(z)}{\sqrt{K_2\tau}} \quad (19)$$

From (9),

$$-K_2 S_2'(z_1) = S_2(z_1) \sqrt{\frac{K_2}{\tau}} = w_{12}[S_1 - S_2(z_1)], \quad (20)$$

or

$$\frac{S_1 - S_2(z_1)}{S_2(z_1)} = \frac{1}{w_{12}} \sqrt{\frac{K_2}{\tau}}. \quad (21)$$

Substituting (17) into (21),

$$\frac{\sigma}{S_2(z_1)} = \frac{C}{w_{12}} \sqrt{\frac{K_2}{\tau}}. \quad (22)$$

Thus we get the predicted $\tau^{-1/2}$ dependency for $\sigma/S_2(z_1)$, and from (1), we find that if τ is given in days (as is the case in the relevant literature),

$$A = \frac{CK_2^{1/2}}{w_{12}} = 2.15C. \quad (23)$$

Jobson et al. (1999) obtained values of A from about 2 to 4 for data sets obtained from lower tropospheric observational studies, which means that our results are in reasonable agreement with the observations for C of order unity.

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