

On the determination of atmospheric minor gases by the method of vanishing partial derivatives with application to CO₂

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[1] We present a general method for the determination of minor gases in the troposphere from high spectral resolution observations. In this method, we make use of a general property of the total differential of multi-variable functions to separate the contributions of each individual minor gas. We have applied this method to derive the mixing ratio of carbon dioxide in the mid-troposphere using data from the Atmospheric Infrared Sounder (AIRS) currently flying on the NASA Aqua Mission. We compare our results to the aircraft flask CO₂ measurements obtained by H. Matsueda et al. over the western Pacific and demonstrate skill in tracking the measured 5 ppmv seasonal variation with an accuracy of 0.43 ± 1.20 ppmv. **Citation:** Chahine, M., C. Barnet, E. T. Olsen, L. Chen, and E. Maddy (2005), On the determination of atmospheric minor gases by the method of vanishing partial derivatives with application to CO₂, *Geophys. Res. Lett.*, *32*, L22803, doi:10.1029/2005GL024165.

1. Introduction

[2] Several fundamental papers using different techniques have recently studied the information content of AIRS spectra. *Aumann et al.* [2005] determined the trend of increase in CO₂ using direct observations from AIRS. *Chédin et al.* [2003], *Crevoisier et al.* [2003], *Engelen and Stephens* [2004] and *Barnet et al.* [2004] have shown that AIRS spectra contain information about trace gases whereas *Engelen et al.* [2004], *Engelen and McNally* [2005] and *Crevoisier et al.* [2004] have retrieved CO₂ from AIRS including seasonal cycle and have compared their results with observations by *Matsueda et al.* [2002]. Recently, *McMillan et al.* [2005] retrieved the distribution of carbon monoxide from AIRS spectra. In this paper, we apply the method of Vanishing Partial Derivatives (VPD) to the AIRS spectra to determine the CO₂ mixing ratio in the middle troposphere and validate the results with the in-situ aircraft flask measurements of Matsueda, taken over the Pacific Ocean at ~10 km altitude between Australia and Japan from Sept. 2002 to March 2004 (flask measurement data are available at <http://gaw.kishou.go.jp/wdceg/pub/index/station/eom9,990.htm#CO2>).

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2. General Approach

[3] We consider the radiative transfer equation

$$R(\nu) = S_s(\nu, \epsilon_s, \dots) + \int_{p_s}^0 B[\nu, T(p)] \left(\frac{\partial \tau(\nu, p, \langle \dots \rangle)}{\partial p} \right) dp \quad (1)$$

where $R(\nu)$, the outgoing radiance at frequency ν measured at the satellite, is the sum of emissions from the surface and the atmosphere. Here ϵ_s is the surface emissivity, B the Planck blackbody function, τ the transmission function from any pressure level p to the top of the atmosphere and the angle bracket $\langle \dots \rangle$ denotes a function of the profiles of temperature $T(p)$, water vapor $q(p)$, ozone $O_3(p)$, carbon dioxide mixing ratio $CO_2(p)$, etc. In this paper, we express the outgoing radiance $R(\nu)$ in brightness temperature units, $\Theta(\nu)$, in order to simplify its use across a wide range of frequencies.

[4] It is well known that the difference between observed radiances and those derived through forward calculations using the retrieved geophysical state of the atmosphere depends on various components including the atmospheric composition. For an iterative method of solution the residual at the n th iteration, $G^{(n)}$ is defined as the sum of squares of the difference between the measured radiances, $\Theta_M(\nu)$, and calculated radiances, $\Theta_C^{(n)}(\nu)$

$$G^{(n)} = \sum_{\nu} [F^{(n)}(\nu)]^2 = \sum_{\nu} [\Theta_M(\nu) - \Theta_C^{(n)}(\nu)]^2 \quad (2)$$

Chahine [1970, Figures 4 and 5] showed that when variable noises, including instrument noise and uncertainty in the CO₂ mixing ratio, are introduced to the simulated spectra the resulting residuals also increase. However, the corresponding error in the retrieved temperature profiles did not systematically increase with this increasing noise level until the RMS noise of all channels exceeds a threshold. In practice we have been able to maintain the accuracy of $1 \text{ }^\circ\text{K km}^{-1}$ for the AIRS retrieved temperature profiles even with an uncertainty in the CO₂ mixing ratio of ± 10 ppmv. For the ideal case when different values of the CO₂ mixing ratio were used to compute $G^{(n)}$, the minimum residual was observed to occur at the true value of the CO₂ mixing ratio, or $\partial G / \partial X \rightarrow \epsilon$, where ϵ is a vanishingly small number.

[5] We now generalize this observation to formulate the problem of determining the concentrations of atmospheric minor gases from remote sounder data. We consider the case where observations are made in a spectral region in the infrared where several gases such as CO₂, O₃, and H₂O are

radiatively active. For simplicity, and without loss of generality, we begin developing the approach for the case where the mixing ratios X_i are constant, independent of pressure. This allows us to consider the function notation $G = G(X_1, X_2, X_3, \dots, X_i)$. In order to find the set of \bar{X}_i which minimizes the residual function G , we express the total differential of G as

$$dG = \frac{\partial G}{\partial X_1} dX_1 + \frac{\partial G}{\partial X_2} dX_2 + \dots + \frac{\partial G}{\partial X_i} dX_i + \varepsilon. \quad (3)$$

Assuming the set of variables ($X_1, X_2, X_3, \dots, X_i$) are linearly independent, the condition that G in equation (3) should have a maximum or a minimum at a point ($\bar{X}_1, \bar{X}_2, \bar{X}_3, \dots, \bar{X}_i$) requires that each of the first partial derivatives

$$\frac{\partial G}{\partial X_1}, \frac{\partial G}{\partial X_2}, \frac{\partial G}{\partial X_3}, \dots, \frac{\partial G}{\partial X_i} \quad (4)$$

should individually vanish at that point.

[6] Thus, we reach the important conclusion that the value of X_i , the mixing ratio of each of the minor gases considered, is that which makes its partial derivative in equation (4) vanish individually. Therefore, even though the observed spectra cannot differentiate between the contributions of individual lines, the partial differentials can. We shall refer to this approach as the method of Vanishing Partial Derivatives (VPD).

[7] We recognize that condition (4) is necessary but we will rely on experience with real data to show uniqueness. Since the variables $X_1, X_2, X_3, \dots, X_i$ are not strictly linearly independent, we adopt an iterative approach and select ‘‘auxiliary sets’’ of channels for T, q, and O_3 to separate the interdependence of the variables in the ‘‘main set’’ which is selected to determine the mixing ratio of the gas under consideration. We should keep in mind that solutions that maximize residuals are also possible but should be rejected.

3. Formulation

[8] In general, the distributions of minor gases are functions of pressure, $X_i(p)$, and thus $G = G(X_i(p))$. We start the iterative solution with initial profiles $X_i^{(0)}(p)$, apply linear transformations $\alpha_i X_i(p)$ to perturb the profiles, where the α_i are scaling coefficients and independent of pressure. We proceed to determine the $\bar{\alpha}_i^{(1)}$ which make the corresponding partial derivatives $\partial G(\alpha_i X_i(p))/\partial \alpha_i$ vanish.

[9] Next, we expand the difference between observed and calculated radiances in equation (2) as a Taylor series and write for any iteration of $n = 1, 2, \dots$

$$F_i^{(n-1)}(\nu, \alpha_i) = a_i^{(n-1)}(\nu) + b_i^{(n-1)}(\nu) \alpha_i \quad (5a)$$

$$a_i^{(n-1)}(\nu) = \left[\Theta_M(\nu) - \Theta_C^{(n-1)}(\nu) \right] \quad (5b)$$

The brightness temperature $\Theta_C^{(n-1)}(\nu)$ is computed from the profiles $X_i^{(n-1)}(p)$, and $b_i^{(n-1)}(\nu)$ is the expansion parameter

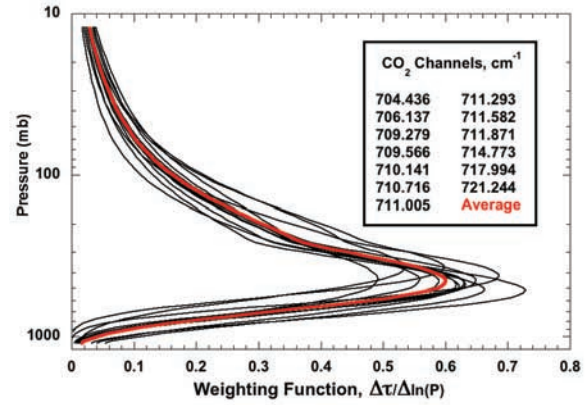


Figure 1. Weighting Functions of the channel set selected to determine the CO₂ mixing ratio. The red curve refers to the average weighting functions for the set.

for each frequency ν , within the small range of variation of α_i . We obtain the sum of squares of the right hand side of equation (5a) separately for the main set and for the auxiliary sets of frequencies and set the derivative of the sum equal to 0. The value of $\bar{\alpha}_i^{(n)}$ which make each term in condition (4) vanish individually is

$$\bar{\alpha}_i^{(n)} = - \frac{\sum_{\nu} a_i^{(n-1)}(\nu) b_i^{(n-1)}(\nu)}{\sum_{\nu} [b_i^{(n-1)}(\nu)]^2} \quad (5c)$$

resulting in new values $X_i^{(n)}(p)$ for each of the variables

$$X_i^{(n)}(p) = \left(1 + \bar{\alpha}_i^{(n)} \right) X_i^{(n-1)}(p) \quad (5d)$$

subject to the condition that $|\bar{\alpha}_i^{(n)}| \ll 1$. In addition we must require that $G^{(n)} < G^{(n-1)}$ to prevent the vanishing partial derivatives in condition (4) from seeking a maximum instead of a minimum.

4. Application to Derive the CO₂ Mixing Ratio from AIRS Observed Spectra

[10] AIRS is a cross-track scanning grating spectrometer with 2378 channels of nominal resolving power of 1200 extending from 3.7 to 15.4 μm with a 13.5 km field of view at nadir [Aumann *et al.*, 2003]. Since its launch in May 2002, AIRS has demonstrated a stability of 10^{-3} $^{\circ}\text{K yr}^{-1}$ with a spectral accuracy of the center frequency of 2 parts per million [Aumann *et al.*, 2004]. The AIRS retrieval algorithm employs information from a companion microwave sounder, the Advanced Microwave Sounding Unit (AMSU) to retrieve T(p), q(p), etc. in the presence of clouds on a horizontal scale of one AMSU field of view or 45×45 km at nadir, the equivalent of 3×3 AIRS footprints [Suskind *et al.*, 2003]. AIRS retrieved temperature profiles $T^{(0)}(p)$ are globally accurate to 1 $^{\circ}\text{K km}^{-1}$ in the troposphere and the retrieved water vapor profiles $q^{(0)}(p)$ are globally accurate to about 15%.

[11] We evaluated the relative sensitivity of channels to CO₂, temperature, water vapor and ozone and chose two

spectral ranges for use in our retrieval. We found the range 690–725 cm^{-1} to be best suited to select the main channel set to retrieve the CO_2 mixing ratio and the two auxiliary channel sets for temperature and O_3 , while the range 1370–1610 cm^{-1} is best suited to select the auxiliary channel set for water vapor. Figure 1 shows the main set of channels selected to retrieve the CO_2 mixing ratio, as well as their corresponding weighting functions, or kernels. The three auxiliary sets are needed to separate the interdependence of temperature, water vapor and ozone on each other and on CO_2 . Specifically, the auxiliary set for temperature is selected from the saturated CO_2 lines, in the same spectral range, to minimize the dependence of temperature on variations in CO_2 while the auxiliary O_3 channel set shows strong dependence on both temperature and CO_2 . It is important to note that the main CO_2 channel set shown in Figure 1 has a strong dependence on temperature but a very weak dependence on water vapor and O_3 . For instance a change of 10% in either the water vapor or O_3 profiles results in a radiance change less than that due to a change of 1 ppmv in CO_2 mixing ratio. We carefully eliminate the effects of uncertainty in surface emissivity and pressure by excluding all channels having a contribution from the surface.

[12] We initiate the iterative solution with the cloud-cleared Level 2 (L2) Retrievals made on a 3×3 AIRS footprint grouping within its associated AMSU footprint, which we will refer to as “AIRS Retrievals”. These data products are available from the GSFC DAAC and can be accessed at http://disc.gsfc.nasa.gov/AIRS/data_access.shtml. The Radiative Transfer Algorithm (RTA) currently used in the AIRS Retrieval assumes a constant CO_2 mixing ratio of 370 ppmv throughout the atmosphere [Strow *et al.*, 2003].

[13] Consider a cluster comprised of J AIRS Retrievals with the following indexing conventions: i is over unknown variables (T , q , O_3 , CO_2), j is over AIRS Retrievals, k is over clusters, and superscript index (n) is the iteration step. Given $[T^{(0)}(p), q^{(0)}(p), O_3^{(0)}(p)]_j$ from each AIRS L2 Retrieval (j) in the cluster, together with its cloud cleared radiances in $\Theta_M(\nu)$ and $\text{CO}_2^{(0)} = 370$ ppmv, we compute the initial ($n = 0$) residuals for the CO_2 sounding channels, $G_{\text{CO}_2}[T^{(0)}, q^{(0)}, O_3^{(0)}, \text{CO}_2^{(0)}]_j$, and proceed to separate the interdependence of our variables as follows, starting with iteration $n = 1$ and AIRS Retrieval $j = 1$:

[14] 1. Using $T^{(0)}, q^{(0)}, O_3^{(0)}$ and $\text{CO}_2^{(n-1)}$, we follow the steps in equations (5) to compute the scaling coefficient $\bar{\alpha}_T^{(n)}$ that will make the partial derivative of the auxiliary temperature channel set vanish, and then calculate the new temperature profile $T^{(n)}$ according to equation (5d). Next, using $T^{(n)}, q^{(0)}, O_3^{(0)}$ and $\text{CO}_2^{(n-1)}$ determine $\bar{\alpha}_q^{(n)}$ and $q^{(n)}$ in the same manner using the auxiliary water vapor channel set. Likewise, using $T^{(n)}, q^{(n)}, O_3^{(0)}$ and $\text{CO}_2^{(n-1)}$ determine $\bar{\alpha}_{O_3}^{(n)}$ and $O_3^{(n)}$ using the auxiliary ozone channel set.

[15] 2. Using $T^{(n)}, q^{(n)}, O_3^{(n)}$ and $\text{CO}_2^{(n-1)}$ determine the scaling coefficient $\bar{\alpha}_{\text{CO}_2}^{(n)}$ that will make the partial derivative of the residuals of CO_2 channel set vanish. Calculate the new mixing ratio of this j th AIRS Retrieval, $[\text{CO}_2^{(n)}]_j$ and compute the corresponding residual of the CO_2 channel set. If $G_{\text{CO}_2}[T^{(n)}, q^{(n)}, O_3^{(n)}, \text{CO}_2^{(n)}]_j < G_{\text{CO}_2}[T^{(0)}, q^{(0)},$

$O_3^{(0)}, \text{CO}_2^{(n-1)}]_j$ is not satisfied the solution for this j th AIRS Retrieval is rejected.

[16] 3. Return to step 1 and repeat the calculation for each of the remaining AIRS Retrievals in the cluster.

[17] 4. Determine the median CO_2 mixing ratio for the cluster. Compute the change in the median, $\Delta^{(n)} = [\text{Median } \text{CO}_2^{(n)} - \text{Median } \text{CO}_2^{(n-1)}]$. If $\Delta^{(n)} < c$, where c is a predetermined limit, a solution for CO_2 has been derived for the cluster. Otherwise, increment n and return to step 1, setting $\text{CO}_2^{(n-1)}$ equal to the median CO_2 mixing ratio derived in this step. This process usually converges within 5 iterations (i.e., $n \leq 5$) for $c = 0.5$ ppmv.

[18] Note that by requiring each iteration to begin in step 1 with the original AIRS retrieved values, $T^{(0)}(p), q^{(0)}(p), O_3^{(0)}(p)$ while using the last derived median CO_2 mixing ratio for the cluster, we are ensuring that the iterative values of CO_2 drive the changes in T, q and O_3 , rather than the contrary.

5. Results

[19] AIRS generates approximately 10^5 AIRS Retrievals daily under various cloud conditions. When comparing AIRS retrieved CO_2 mixing ratio with Matsueda flask measurements, a cluster is defined as a set of AIRS Retrievals that are all collocated with a Matsueda flask measurement in time and space. Our collocation requirement is that each AIRS Retrieval must occur within a radius of 150 km and within ± 4 hours of the flask measurement. Of the 402 flask measurements posted by Matsueda *et al.* there are 223 with which one or more AIRS Retrievals are collocated. We identify a total of 2332 AIRS Retrievals collocated with the 223 Matsueda measurements (i.e., 233 clusters, one per Matsueda measurement). This large number of collocated AIRS Retrievals would be reduced by one order of magnitude if only “nearly cloud free” retrievals are selected.

[20] Since we use AIRS Retrievals made in the presence of cloud, we base our answer in step 4 on the cluster median value of CO_2 to minimize the impact of extreme outliers. Subsequently, we exclude from consideration any of the 223 clusters that contain less than three AIRS Retrievals and reject all AIRS Retrievals that do not seek a minimum during the iteration process. As a result, the 223 clusters and collocated Matsueda measurements are reduced to 103 containing 927 AIRS Retrievals, for an average of $J = 9$ acceptable AIRS Retrievals per cluster.

[21] Matsueda measurements exhibit seasonal variations in CO_2 of about 5 ppmv during the period of comparison, as shown in Figure 2. Comparison of the 103 individual AIRS clusters to their collocated Matsueda measurements result in a Bias (Matsueda-AIRS) = 1.15 ppmv, with a Standard Deviation (SD) of ± 3.1 ppmv, demonstrating AIRS ability to track the 5 ppmv seasonal variation of Matsueda with a SD of ± 3.1 ppmv. For the monthly statistics shown in Table 1 there are about 7 clusters per month over the 14-month period. We select the monthly median value from the AIRS CO_2 results and compare each to the corresponding monthly median of the Matsueda measurements. The monthly statistics of [Matsueda-AIRS] for the 14 months result in a SD = ± 1.37 ppmv. (When

we compare monthly means instead of medians the corresponding SD increases slightly to ± 1.47 ppmv).

[22] Next, we apply an internal quality check to improve our results. We observe that the average SD of the 103 clusters of AIRS Retrievals is 1.12 ppmv and that some clusters have a SD > 2.0 ppmv. By excluding clusters with SD > 2.0 ppmv we reduce the number to 78 clusters, containing 731 AIRS Retrievals, still with an average of 9 AIRS Retrievals per cluster. The resulting 78 cluster comparison with their collocated Matsueda measurements in Table 1 shows a Bias = 1.01 ppmv, and SD = ± 2.98 ppmv and the corresponding monthly statistics yield SD = ± 1.20 ppmv. Thus, we conclude that the combination of averaging, use of median values together with a quality checks leads to the best tracking results that we show in Figure 2.

[23] What is even more revealing on the performance of the VPD method is that the distribution of the individual [Matsueda-AIRS] tracking errors exhibits Gaussian properties. The computed SD of [Matsueda-AIRS] for the 78 individual collocations and that for monthly averages of 7 clusters per month are approximately related by $2.98/\sqrt{7} = 1.13$. Thus, the use of averaging should improve the results of the derived CO₂ distribution.

[24] The small magnitude of the standard error of the mean, shown as vertical bars on Figure 2, is partly due to the very high stability of AIRS data and to the 1°K km^{-1} accuracy of the temperature profiles and the robustness of the VPD method. By starting each iteration with a “new derived” CO₂ mixing ratio but with the original AIRS retrieved geophysical parameters we have assured a more coherent results. Overall, the requirement that the residuals must continue to diminish in each iteration is a very powerful intrinsic condition that determines which AIRS

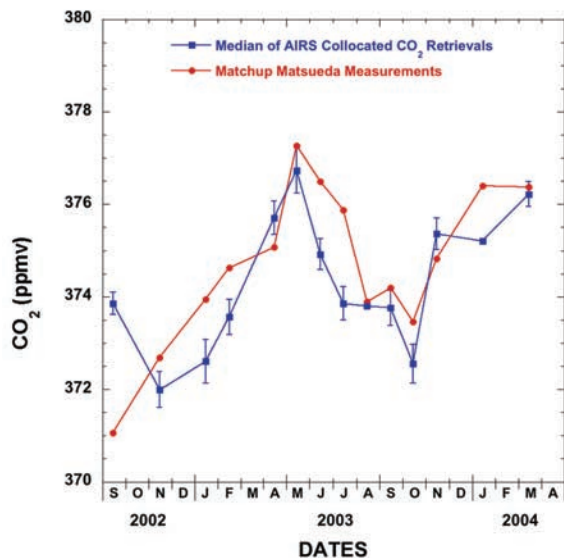


Figure 2. Comparison of AIRS retrieved CO₂ mixing ratio with aircraft flask measurements by Matsueda et al., showing skill in tracking the measured seasonal variation of 5 ppmv with an accuracy of 0.43 ± 1.20 ppmv. The monthly error bars are the standard error of the mean.

Table 1. Comparison of Matsueda and AIRS^a

Number of. Match-ups per AIRS Clusters	[M-A] Bias	[M-A] SD
103 individual ^b	1.15	3.10
14 Months ^c	0.63	1.37
78 individual ^d	1.01	2.98
14 Months ^e	0.43	1.20

^aIn units of ppmv. SD, standard deviation; M, Matsueda (the truth); and A, AIRS results.

^bNumber of clusters with 3 or more AIRS retrievals.

^cAverages of 7 clusters/month.

^dNumber of clusters with a SD < 2 ppmv.

^eAverages of 6 clusters/month.

Retrievals should be accepted and which ones should be rejected.

6. Future Applications

[25] We have demonstrated that accurate CO₂ mixing ratios can be derived from infrared observations made in the presence of clouds. These results are significant for future applications to determine the global distribution of CO₂. The Gaussian properties of tracking errors indicates that averaging over time and space should yield improved accuracy of the derived CO₂ distribution.

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