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Atmospheric Measurement Techniques Discussions

High-resolution air quality monitoring from space: a fast retrieval scheme for CO from hyperspectral infrared measurements

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Abstract

The first results of the Fast Linear Inversion Trace gas System (FLITS) retrieval scheme are presented here for CO from IASI (Infrared Atmospheric Sounding Interferometer) measurements using RAQMS (Real time Air Quality Modelling System) as atmospheric background. FLITS is a simple linear inversion scheme with a stable performance that retrieves total column CO concentrations (molec cm^{-2}) at single field-of-view (FOV) irrespective of cloud cover. A case study is presented here for a biomass burning plume over the Pacific on 29 March 2010. For each FOV a single tropospheric CO density, vertically integrated over 200–800 hPa, is retrieved with 12 channels in the spectral range 2050–2225 cm^{-1}. Despite variations in cloud cover and temperature, the degrees of freedom for signal (DFS) of the solution ranges between 0.8 and 0.95. In addition, the retrieval error is at least half the background error of 10 %, with dominant contribution from uncertainty in the measurement and temperature. With its stability and processing speed, FLITS meet two of the key requirements for operational processing. We conclude that the linear combination of space-borne measurements with a chemical transport model in the FLITS retrieval scheme holds potential for real-time air quality monitoring and evaluation of pollutant transport at high spatial resolution.

1 Introduction

The study of complex environmental systems, such as the atmosphere, relies on a time series of accurate measurements at a range of spatial scales. Since the first measurements of the atmosphere, one of the main objectives has been to develop a predictive understanding of its behavior and processes, especially as it impacts society. Much effort has gone into numerical weather prediction and climate modeling with most of the first five decades of space-borne measurements dedicated to improved predictive capability. It is only in recent years, with the growing awareness of pollution and its

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impact on human health, that effort has gone into measuring trace gas species from space with the goal to study global atmospheric chemistry and transport.

Where most of the instruments launched on spacecraft have been designed to measure temperature (T), water vapor (H_2O), clouds and surface properties, a number of recent instruments have the capability to specifically measure trace gas species, e.g., SCHIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Cartography), MOPITT (Measurements Of Pollution In The Troposphere), TES (Tropospheric Emission Spectrometer), AIRS (Atmospheric InfraRed Sounder) and IASI (Infrared Atmospheric Sounding Interferometer). We are now seeing the routine retrieval of dominant greenhouse gases daily on a global scale, O_3, CO_2, CH_4 and CO (e.g., Bergamaschi et al., 2000; Clerbaux et al., 2003; Martin, 2008; Razavi et al., 2009; Boynard et al., 2009), and experimental work on HNO_3 (Wespes et al., 2009), SO_2 (Clarisse et al., 2009) as well as reactive species such as ethane, methanol and formic acid in fire plumes (Coheur et al., 2009). In this paper, we are interested in CO that absorbs infrared radiation in the range 2050–2225 cm^{-1}.

Most of the reported success has been achieved for retrievals spatially averaged to a resolution lower than what is measured by the instrument for improved accuracy and stability. This is as much a result of instrument limitation as it is of the science objectives and retrieval algorithms. Pollution transport can only be understood as a system once it is measured repeatedly on multiple spatial scales. Networks of point-source measurements give accurate trace gas counts but with a limited spatial range and a bias toward industrial zones. Space-borne retrievals of atmospheric parameters give global trace gas distributions with reasonable accuracy but at a low spatial resolution. There is a data shortage between the highly localized and the globally averaged and a need to measure pollution events on a moderate to high spatial resolution.

The reason for this shortage can be explained when one considers the difficulty with which to detect weak absorbers of outgoing radiation from space. A number of studies have measured the information content (IC) of top of atmosphere (TOA) measurements with respect to the major pollutant gases (e.g., Rodgers and Connor, 2003; Engelen and Stephens, 2004) and found them to be considerably lower than what is measured for T and H_2O, both treated as strong absorbers in retrieval systems. George et al. (2009), for example, reported the IC (calculated as the degrees of freedom for signal, DFS) of IASI with respect to CO to vary globally between 0.8 and 2.4. Turquety et al. (2009) similarly measured a DFS of 1.6–1.8 for CO over the Mediterranean. In other words, the most advanced infrared sounder in space today (i.e., IASI) with more than 8000 instantaneous spectral measurements has at most 2.4 independent pieces of information about the vertical structure of CO. What is more is that George et al. (2009) reported this sensitivity to CO to be concentrated mostly in the mid troposphere. This is in stark contrast with similar reports of IASI with respect to T, which showed sensitivity peaks at levels throughout the atmospheric column (from 0–60 km) and a DFS well above 10 in most cases (Lerner et al., 2002).

A popular method for processing TOA measurements with a low IC is to retrieve the target species as a profile at a number of levels (nlev) higher than what is suggested by the DFS. While compromising the signal-to-noise ratio (*S/N*), it allows a full vertical description of the atmosphere. When the information retrieved exceeds the information available in the measurements (i.e., nlev > DFS) it is referred to as an under-determined inversion problem. The *S/N* can be improved in a solution by applying appropriate constraints that include, defining an a priori for the retrieval parameter and iterating through a series of solutions until a pre-defined cost function (or error threshold) is minimized. Rodgers (2000) describes this as Optimal Estimation (OE) and today it is one of the most popular approaches used in the retrieval of atmospheric parameters, particularly trace gases (Barret et al., 2005; Coheur et al., 2009; Illingworth et al., 2010). We use the term trace gas to include all major atmospheric gasses with the exception of H_2O. Their sparse concentration in the atmosphere makes them weak absorbers of outgoing radiation, which renders their retrieval from TOA radiances highly non-linear. The popularity of OE stems from the fact that non-linearity is partly addressed in the iterative assessment of a cost function.

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In contrast to these, we illustrate here that when a good background estimate of the atmosphere is available, the inversion of IASI with respect to CO can be treated as a simple linear problem. We are specifically interested in how this applies to operational environments of data processing and the monitoring of atmospheric conditions in real-time. OE retrieval systems are computationally complex and in resource-poor environments can be very slow. A linear system, on the other hand, offers advantages in terms of processing speed and computational simplicity since inversion is performed in a single step.

A full description (i.e., T, H_2O and all the major gases) of the atmosphere is provided by the global trace gas chemical transport model, Real-time Air Quality Modelling System (RAQMS) (Pierce et al., 2007). We use it here as background estimate in the linear trace gas retrieval scheme, FLITS (Fast Linear Inversion Trace gas System) (Smith, 2010). The latter is a fast and intuitive method that retrieves CO as a total column density from IASI soundings. In this paper, the first results of the FLITS scheme are presented for a biomass burning event over the Pacific Ocean on 29 March 2010. With it we demonstrate the stability that can be achieved with a linear retrieval system despite the inherent non-linearity of the problem. We argue that the processing speed and stability of FLITS make it a strong candidate for implementation in an operational environment. Routine retrievals can be performed twice daily (once for every overpass of the Metop-A platform) to map CO patterns and transport at instrument resolution (12 km for IASI). RAQMS provide predictive measurements four times a day but at a low spatial resolution. FLITS retrievals compliment RAQMS by adding spatial structure, highlighting certain features otherwise unknown. Unlike the IASI L2 processor, no spatial averaging is performed on FLITS retrievals.

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2 Methods and data

2.1 Total column inversion algorithm

The FLITS retrieval scheme is set within a statistical Bayesian framework in which the knowledge of each variable is given in terms of its probability density function (PDF) that has a sample mean and variance. Rogers (2000) give a full description of this framework as it applies to atmospheric sounding. In short, it allows the inversion of the measurement PDF at the point of minimization with the atmospheric background PDF. The retrieval is itself a PDF but its mean, also known as the maximum likelihood (ML) value, is selected as final solution with the variance an indication of the uncertainty associated with the mean.

A FLITS retrieval is performed in two steps. First, the fraction (or percentage) change in the background estimate of the parameter is retrieved from the measurements as:

$$\delta \mathbf{x} = \frac{\mathbf{k}^T \mathbf{S}_\varepsilon^{-1} (\mathbf{y} - \mathbf{y}_a)}{(\mathbf{k}^T \mathbf{S}_\varepsilon^{-1} \mathbf{k} + 1/\mathbf{s}_a)}. \tag{1}$$

where $\mathbf{y}$ is the space-borne measurement (1 × number of channels (nchan)) of TOA radiance, $\mathbf{y}_a$ the background TOA radiance (1 × nchan), $\mathbf{s}_a$ (1 × 1) the background error and $\mathbf{S}_\varepsilon$ (nchan × nchan) the instrument noise covariance matrix, $\mathbf{k}$ (1 × nchan) the weighting function vector that describes the sensitivity of $\mathbf{y}_a$ with respect to $\mathbf{x}_a$. The weighting function is calculated here as $\mathbf{k} = (f(\mathbf{x}_1) - f(\mathbf{x}_2))/0.1$ with $\mathbf{x}_2 = 0.9\mathbf{x}_1$. It gives the change in $\mathbf{y}$ given a 10 % change in $\mathbf{x}$. The shape and magnitude of $\mathbf{k}$ determines the shape and magnitude of the analysis increments (or retrieval parameter corrections) of the background estimate. In order to understand the dependence of a retrieval on the a priori, Eq. (1) can be rewritten as $\delta \mathbf{x} = (\hat{\mathbf{x}} - \mathbf{x}_a)/\mathbf{x}_a$, where $\hat{\mathbf{x}}$ is the retrieval- and $\mathbf{x}_a$ the a priori parameter. This leads to the second step in which a CO total column estimate is reconstructed by adding the a priori to the fraction retrieved in step 1:

$$\hat{\mathbf{x}} = (1 + \delta \mathbf{x}) \mathbf{x}_a. \tag{2}$$

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The retrieval parameter $\hat{\mathbf{x}}$ is a tropospheric total column density value (molec cm^{-2}) vertically integrated over the range of maximum sensitivity to CO, 200–800 hPa. This vertical integration is achieved in two of the algorithm variables namely, (i) in the calculation of $\mathbf{k}$, the a priori profile is perturbed at once along the full range, thereby treating the troposphere as a single thick layer, and (ii) before the reconstruction of $\hat{\mathbf{x}}$, the mixing ratio profile $\mathbf{x}_a$ is integrated to column density along this range.

In the event that a highly non-linear problem is linearized, the solution can be very unstable since the complexity of error source and propagation is not explicitly addressed. However, degrees of stability are introduced in FLITS through careful channel selection, definition of the atmospheric background and surface parameters, and adding surface temperature (ST) as a second, or joint, retrieval parameter. The latter is achieved by adding a second layer the weighting function vector, $\mathbf{k}$ ($2 \times$ nchan) and background error value, $\mathbf{s}_a$ (2×2) in Eq. (1).

A subset of 12 channels is selected individually for each retrieval. They are selected in the spectral range 2050–2225 cm^{-1} in pairs of three along the main CO absorption lines with a method described as Absorption Line Cluster (ALC) channel selection (Smith, 2010). In the definition of the instrument noise, $\mathbf{S}_\varepsilon$, a correlation factor of 0.71 is assumed between adjacent channels with the square of the instrument noise equivalent delta temperature (NEdT) on the diagonal. The magnitude of this correlation factor is a common assumption for adjacent IASI channels in atmospheric retrievals (Lerner et al., 2002). A background error of 10 % was set for CO in accordance with the expected accuracy of global CO retrievals from IASI, with 0.5 K for ST.

Before the CO tropospheric densities are reconstructed in Eq. (2), a quality filter is applied to remove all cases where $\delta\mathbf{x} > 1$. In other words, when the retrieval replaces the a priori, as happens when the fraction change retrieved is more than twice the value of the a priori, then the retrieval is considered to be invalid.

In Sect. 3, we use two measures with which to evaluate the FLITS retrieval (both are discussed in detail by Rodgers, 2000). The first is the averaging kernel, $\mathbf{A} = (\mathbf{k}^T \mathbf{S}_\varepsilon^{-1} \mathbf{k} + \mathbf{s}_a^{-1})^{-1} \mathbf{k} \mathbf{S}_\varepsilon^{-1} \mathbf{k}$. It quantifies the sensitivity of the retrieval to the true value as a fraction between 0 and 1. A kernel of unity indicates that the inversion is independent of the a priori with all information derived from the TOA measurements. Similarly, a kernel of 0 indicates a reproduction of the a priori in the retrieval. Usually its value ranges between 0 and 1 and thus communicates the degree of contribution from each source in the solution; the ML value is often described as a weighted mean. Given the FLITS retrieval dimensions the kernel is a scalar value but we maintain reference to **A** (instead of *a*) to avoid confusion. The averaging kernel can also be interpreted as an IC measure since:

$$\mathrm{DFS} = \sum_{i=1}^{\mathrm{nlev}} \mathbf{A}_{ii} \tag{3}$$

In the FLITS retrieval system the kernel is already vertically integrated, thus it follows that DFS = **A**. The second measure used in the evaluation of FLITS retrievals is the retrieval error:

$$\hat{\mathbf{s}} = (\mathbf{A}-1)\mathbf{s}_a\,(\mathbf{A}-1) + \mathbf{G}\mathbf{S}_\varepsilon\mathbf{G}, \tag{4}$$

where **G** is the gain function, $\mathbf{G} = (\mathbf{k}^T\mathbf{S}_\varepsilon^{-1}\mathbf{k} + \mathbf{s}_a^{-1})^{-1}\mathbf{k}^T\mathbf{S}_\varepsilon^{-1}$. The first term on the right-hand side in Eq. (4) is known as the smoothing error. It can be described as the information loss due to a lack of sensitivity to the true state. The second term, the measurement error, describes the combined error due to instrument noise, uncertainty associated with the forward model, background parameters not directly retrieved (e.g., T and H_2O) as well as correlation among neighboring channels.

2.2 Background atmospheric state

Being a linear retrieval scheme, the FLITS solution is highly dependent on a background estimate ($\mathbf{x}_a$, $\mathbf{y}_a$) because the inversion of $\mathbf{y}$ with respect to $\hat{\mathbf{x}}$ is achieved in a single step. A full definition of the atmospheric state at a given time and space is required in the simulation of $\mathbf{y}_a$ (Eq. 1) and a total column CO value is required for the

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a priori parameter, $\mathbf{x}_a$ (Eq. 2). We used modeled fields to make up the full atmospheric definition.

RAQMS provides a full thermodynamic description of the atmosphere (T and H_2O) together with a chemical description of the trace gas species CH_4, O_3, N_2O and CO. (CO_2 is defined as a constant profile ranging from 365 ppmv at TOA to 375 ppmv at the surface.) It produces these fields daily at six hour intervals (starting at 00:00:00) on a spatial grid of 2° × 2°. A full description of the RAQMS model is available in a recent publication (Pierce et al., 2007). Each RAQMS profile is defined along a unique 35 level vertical grid with its surface pressure set at the topographic elevation of the grid point geographic coordinate. The RAQMS CO values have been validated against those retrieved from MOPITT and both are in good agreement (Pierce et al., 2007), albeit with the former displaying up to a 10 % positive bias.

RAQMS lack a description for two surface parameters, ST and emissivity. In an analysis of spectral sensitivity, it is apparent that IASI radiances are sensitive to both parameters in the CO absorption range. It is, thus, important that they be defined with high accuracy. In the definition of ST we contended with using the definition for boundary layer temperature (provided by RAQMS) in this paper since ocean ST has a low variability compared to land and the retrieval vertical range exclude the surface by extending only up to 800 hPa with the ocean surface usually at 1100 hPa.

Surface emissivity, on the other hand, was defined by the global database of infrared surface emissivity as developed by UW-Madison (Seeman et al., 2008). These emissivity values are based on the MODIS (Moderate Resolution Imaging Spectrometer) operational land surface emissivity product (MOD11) that has emissivities defined along six points in three spectral regions; 3.8–4, 8.6 and 11–12 µm. However, CO absorption range centers on 4.7 µm. The UW baseline fit method that is trained on laboratory measurements, redefines the MOD11 product along ten spectral points (3.6, 4.3, 5.0, 5.8, 7.6, 8.3, 9.3, 10.8, 12.1 and 14.3 µm) and thus better captures the emissivity spectrum along the full infrared range. We interpolated the UW baseline fit emissivity spectra to the IASI spectral resolution for use in the FLITS scheme.

The FLITS retrieval is performed at IASI single FOV resolution. Thus, where necessary the data were linearly interpolated to the IASI L1C latitude/longitude grid. In addition, the RAQMS fields were interpolated to fit the temporal definition for each IASI granule.

2.3 IASI radiances

The IASI instrument onboard the European MetOp-A satellite was launched on 19 October 2006. It is the first of three successive infrared sounders scheduled for launch into low-Earth orbit for global operational meteorology. With its apodized spectral resolution of 0.5 cm^{-1} (spectral sampling of 0.25 cm^{-1}) it has 8461 measurements in the infrared range, 645–2760 cm^{-1}. This together with its low radiometric noise (on-flight analysis puts its NEdT at a range between 0.06 and 0.4 K) and improved horizontal coverage (it has an across track swath width of 2200 km) it offers some of the most advanced atmospheric measurements with a global coverage twice daily at 12 km footprint size. The reader should refer to Clerbaux et al. (2009) for an updated discussion of the IASI operational performance. IASI Level 1C (L1C) radiances are available online (https://class.ncdc.noaa.gov).

The background estimate IASI radiances at TOA ($\mathbf{y}_a$) are simulated with the radiative transfer model, RTTOV version 9.3 (Matricardi et al., 2004). RTTOV-9.3 is a fast radiative transfer model and uses profile-dependent predictors to parameterize the atmospheric optical depths. This makes it fast enough for near real-time processing. Although it does not calculate line-by-line radiances, the resultant spectra have accuracies at or below instrument noise.

3 Results and discussion

The FLITS retrieval was performed on 12 IASI L1C granules of the descending orbit on 29 March 2010 (18:26:53 to 20:23:57) over a biomass burning plume in the Pacific

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Ocean. No distinction was made between cloudy and clear-sky cases and retrievals were performed on all FOVs. The RAQMS CO a priori, FLITS retrieval product, IASI L2 CO product (http://class.ngdc.noaa.gov) and MOPITT L3 daily gridded CO product (ftp://l4ftl01.larc.nasa.gov) are each plotted for the study region in Fig. 1.

The FLITS quality filter removed retrievals mostly in thick clouds. We distinguish clouds by evaluating the window channel (900 cm^{-1}) brightness temperature (BT) in each FOV. When the BT is significantly lower than surrounding background values, then the FOV is considered to be cloudy. Fewer retrievals are removed this way than would be by a cloud mask. This is valuable for visualization and qualitative analysis of CO horizontal transport irrespective of cloud cover. When the FLITS retrieval results are compared with the operational IASI L2 CO product (Fig. 1b and d), similar circulation patterns are observed, some of which are absent in the RAQMS a priori (Fig. 1a). This illustrates the strength of the retrieval, which we expect to improve over land when a good estimate of the surface temperature is available. (IASI sensitivity to CO is positively correlated with ST.) On average the FLITS retrievals are 1.0 exp 18 molec cm^{-2} higher than the IASI L2 product. This is largely due to biases in the a priori and low sensitivity to CO over ocean.

In the absence of accurate high resolution validation data, comparisons with CO retrievals from other operational nadir looking infrared instruments are common, with MOPITT a popular choice (Warner et al., 2007; Yurganov et al., 2008; George et al., 2009). However, differences in spatial coverage and orbital mechanics render their daily comparison nearly impossible. All three studies referenced here compared different instrument CO retrievals on a global scale for at least a month of data. The MOPITT CO product coincident with our FLITS retrievals is plotted in Fig. 1 to illustrate this point.

We regard a retrieval system as stable when it yields an averaging kernel with a low standard deviation, irrespective of the mean kernel value. A linear retrieval is a weighted mean of the a priori and measurement. Thus, a stable system will yield retrievals with a consistent dependence on the measurement and a priori, respectively.

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This makes it possible to define a background and instrument error once off, without adjustment between retrievals.

In the case presented here, irrespective of the FOV BT, the FLITS kernel ranges between 0.8 and 0.95 in the total column troposphere (200–800 hPa). This can be interpreted as the measurements contributing 80–95 % of the information retrieved. It is a remarkably stable result for a linear retrieval scheme that can easily range between 0.01 and 0.95 across the FOVs in a single IASI granule. Much of this is attributable to the channel selection. The ALC channel selection method used in FLITS is designed to maximize the DFS in a retrieval system. Other objectives for channel selection may include minimum interference from coincident absorbers (as is the case with the IASI operational L2 CO channels set; Turquety et al., 2004) or optimization of CO sensitivity as expressed by the *S/N* (Barnet et al., 2004). A high DFS is necessary in FLITS retrievals to approximate a linear inversion problem. In addition to careful channel selection, ST is added as a joint retrieval parameter to reduce error propagation due to uncertainty in the ST estimate, which we know is sub-standard. In the same way other coincident absorbers can be added as joint retrieval parameters, e.g., H_2O, N_2O, O_3 and T. However, this will greatly increase the dimensions of the weighting function and background error matrices in Eq. (1) and compromise our objective of a simple and fast retrieval system.

The averaging kernel can be converted into DFS (Eq. 3) to communicate the total vertical IC of the retrieval parameter in the measurements. The IASI DFS for CO ranges globally between 0.8 and 2.4 but the DFS in a FLITS retrieval can never exceed unity because DFS $\leq$ nlev by definition, and in FLITS a single CO level is retrieved, i.e., nlev = 1. Thus, care should be taken not to interpret the DFS communicated here as an indicator of the total amount of CO information available in IASI measurements. The DFS reported for FLITS retrievals is close enough to unity to render the inversion problem nearly linear with a low degree of under-determinancy.

The retrieval error (Eq. 4) is on average 5 % for all granules, which is significantly lower than the background error of 10 %. The dominant contribution to the retrieval

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error, $\hat{\mathbf{s}}$, is the measurement error with an insignificant contribution from the smoothing error. This can be traced back to the averaging kernel that indicates the dominant information source. The consistently high kernels in FLITS retrievals makes it possible to conclude that smoothing will rarely cause a significant information loss and that uncertainty in the atmospheric background T and H_2O as well as the forward model and instrument noise will dominate $\hat{\mathbf{s}}$. In highly under-determined retrieval schemes, kernel values around 0.1 are routinely reported for IASI retrievals in tropospheric layers, which means that smoothing, or lack of sensitivity to the real state, makes a significant error contribution. Barret et al. (2005) recorded CO kernels up to 0.3 in tropospheric layers for the Interferometer Monitor of Greenhouse gases (IMG) instrument and George et al. (2009) recorded CO kernels between 0.1 and 0.6 for IASI, AIRS, MOPITT and TES, respectively. Any alteration in the design of a retrieval system (e.g., instrument choice, channel selection, weighting of the error terms, definition of atmospheric layers, etc.) directly affects the averaging kernel and by implication the structure of the retrieval noise.

In this paper, we demonstrated that our approach to linearly combine a chemical transport model as atmospheric background with space-borne infrared measurements yield CO total column densities at an improved spatial resolution compared to the global operational IASI product. Others have attempted high resolution IASI CO retrievals over specific biomass burning sites, but not without spatial averaging (Turquety et al., 2009). FLITS is a simple algorithm that requires no pre-processing (e.g., cloud clearing), cloud masking, iteration through a cost function, or error adjustments. It allows the retrieval of CO distribution patterns with reasonable precision at high spatial resolution that can give valuable insight into pollution transport and air quality processes. The FLITS retrieval system is evaluated in more detail in a follow-up paper with full characterization, error analysis and sensitivity studies, but we conclude here that FLITS holds potential for implementation in a real-time environment where speed of processing and stability is critical.

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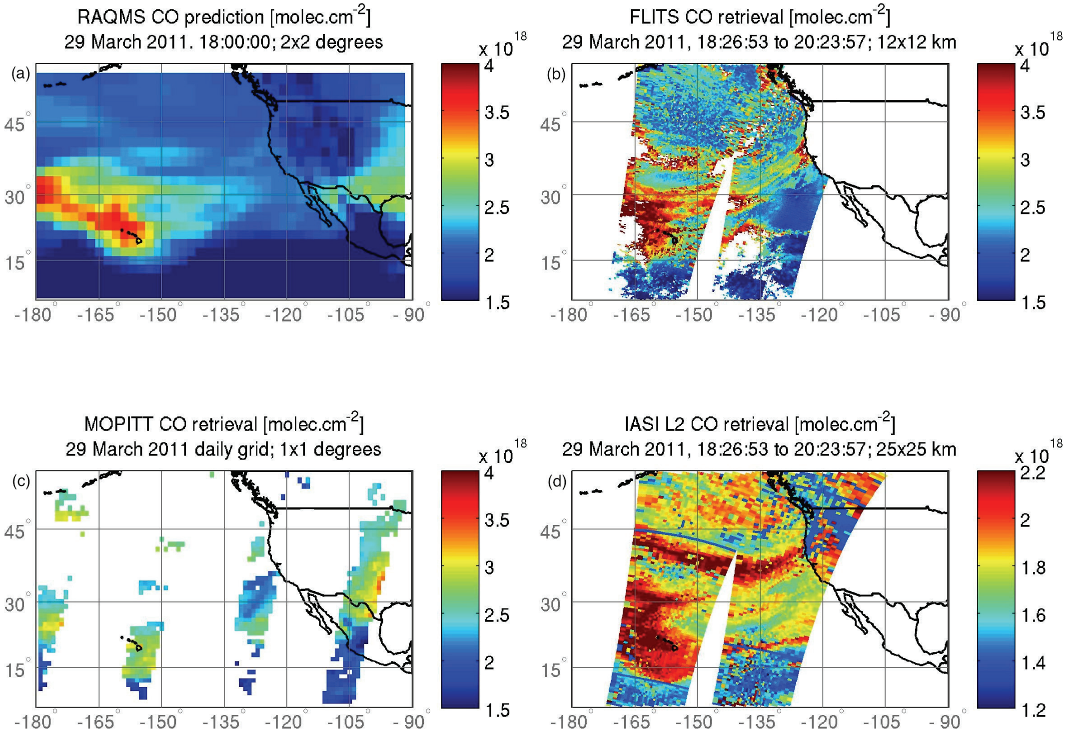

Fig. 1. Total column (200–800 hPa) CO densities on 29 March 2010, **(a)** RAQMS CO prediction, **(b)** FLITS CO retrieval, **(c)** MOPITT Level 3 (version 4) daily gridded CO product, **(d)** IASI Level 2 operational CO product.

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