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Interactive comment on "Remote sensing CO_2, CH_4 and CO emissions in a polluted urban environment" *by* Denis M. O'Brien et al.

Denis M. O'Brien et al.

denis.obrien@ggma.com.au

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The reviewer's comments are shown below in italics, while our responses are in normal font. Revised text is quoted (indented).

General Comments

General comment 1

Section 3.3 : For the later discussion wrt SNR and retrieval sensitivities it is important

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to know what surface albedos (as a function of wavelength) have been used. Please provide that info (incl. ref.), preferably in terms of a map(s) similar to Fig. 3. Also the link between what albedo and SZA (SZA could fixed to some representative value if need be) combinations correspond to which SNR as presented in Fig. 11 is needed.

The text clearly states on page 6, line 9 of section 3.3 that, when simulating the Stokes vector at the top of the atmosphere, '[s]urface properties, such as polarized bidirectional reflectance distribution function (from MODIS and POLDER), were assigned and interpolated to the frequencies of the geoCARB spectral bands'. As explained by O'Dell et al. (2012), the retrieval algorithm assumes instead that the surface is Lambertian with albedo varying linearly across each of the geoCARB bands. The slope and offset are retrieved parameters for each band. Our manuscript refers to O'Dell et al. (2012) for details of the basic retrieval algorithm.

Polonsky et al. (2014), cited in section 3.3, provides in his Figure 4 a simplified model for the signal-to-noise ratio as a function of $A \cos \theta_s$ for the four geoCARB bands, where A is the surface albedo and θ_s is the solar zenith angle.

Because this material has been presented previously, we do not think it need be repeated here. Nevertheless, in order that the spectral interpolations are clearly stated, we have replaced the single paragraph in section 3.3 with the following.

> Surface properties, such as polarized bidirectional reflectance distribution function (BRDF, derived from MODIS and POLDER), were interpolated to the wavelengths of the geoCARB spectral bands. The method was similar to that described by O'Brien et al. (2009) and Polonsky et al. (2014). In the geoCARB spectral bands at 765 nm, 1606 nm and 2065 nm, the surface BRDF was interpolated linearly from the MODIS BRDF at 645 nm, 859 nm, 1240 nm, 1640 nm and 2130 nm. The BRDF in the geoCARB band at 2323 nm was assumed to be the same as the MODIS BRDF at 2130 nm,

because MODIS BRDF is not available at longer wavelengths. The polarised component of the BRDF was assumed to be independent of wavelength. Surface heights from the WRF-Chem topography were interpolated to the centres of the geoCARB pixels.

General comment 2

Section 5.2 prior aerosols : I did not understand the adjusted aerosol retrieval approach. Please describe what you are effectively doing.

The steps of the adjustment are clearly and precisely described by the current text in the paragraphs beginning at lines 16 and 29 of page 9 of the section titled 'Prior aerosol'. The reviewer appears to seeking a simpler explanation. The essential point of the adjustment, that we are trying to model the wavelength dependence of the extinction coefficient more appropriately for the region, is stated briefly in the sentence beginning at line 16 of page 9. We have replaced the opening sentences of the paragraph beginning at line 16 on page 9 with the following.

> Although aerosol loadings can be highly variable in space and time, in the absence of other data it is common to assume that the aerosol type is the same over a limited region. Of the extinction coefficient, single scattering albedo and phase matrix, the most important to capture reliably for the retrieval algorithm is the wavelength dependence of the extinction coefficient. Therefore, after inversion of the simulated spectra with the standard algorithm, the inversions were repeated with the wavelength dependence of the aerosol extinction coefficients adjusted as follows to represent the regional aerosols more accurately.

General comment 3

Section 7.1 p. 12 it is suggested that the problems encountered with the CO retrievals could be alleviated and that a code to do that is available (p.12,l.23). So why was that not done/used ?

As the present text states, there is code available for constrained optimisation. However, incorporating that code in the retrieval algorithm is a major task going far beyond the scope of the present paper. In this paper, we investigated the feasibility of retrieving column-averaged concentrations of X_{CO_2}, X_{CH_4} and X_{CO} in situations with high aerosol loadings, such as Shanghai. We used the tools available, including the ACOS retrieval algorithm. Our study has revealed a deficiency of the algorithm, and we can see a path ahead using existing constrained optimisation tools to improve the algorithm. Following that path is a significant undertaking, and we will report the results in due course.

General comment 4

At the end of section 7.2 suddenly the result of other (?) error sources (other than instrument measurement noise) are mentioned leading to the provided 'actual error'. The background as to what error sources have been used here and how large these were assumed (and their functional form) remains unclear and is not explained. Only some general remarks are made (imperfect spectroscopy, incorrect optical properties aerosols). This is very unsatisfactory as in practice these errors could be dominating and are often systematic. Please elaborate on what was assumed here and what simulations were done which resulted in the errors shown in Fig. 11. (In the end it was not clear to me if only instrument meas. noise was taken into account or also other error sources in Fig. 11.)

These issues have been dealt with comprehensively in the extensive literature on re-

trieving trace gas concentrations from spectra of reflected sunlight. Therefore, we do not think there is any need to elaborate in this paper. As stated in many papers, including this paper in equation (5) and references cited therein, Bayesian inversion requires not only the covariance of the instrument noise but also the prior error covariance and the jacobian that maps state variables to spectra. The easiest component to specify is the instrument noise, while the jacobian is the most difficult, because it depends on many parameters of the system comprised by atmosphere, surface and instrument. A short list would include cloud and aerosol optical properties, spatial inhomogeneity and variations of surface reflectance over the scene,

The contributions to error from the jacobian are difficult to quantify for obvious reasons. One commonly accepted methodology for studying the consequences of such errors is through numerical experiments in which an ensemble of spectra at the top of the atmosphere, generated using a detailed simulator of the atmosphere, surface and instrument, are analysed with an algorithm to which the detailed information is denied. That is the approach followed in this paper. As all this is well known and documented in the literature, we reject the reviewer's assertions.

General comment 5

Section 8 p.14,l.22 : concentration retrieval errors that can be expected from a ***real*** *instrument. It remains unclear to me to what extent the full instrument behavior is simulated other than the instrum. meas. noise error. Because if only instrum. meas. noise is accounted for in the errors presented here, it will only a be a lower limit to the real errors.*

Indeed there are instrument uncertainties that we have not modelled, such as those associated with radiometric calibration and pre-flight measurement of the instrument line shape function. However, published studies (for example that by O'Dell et

al. (2012)) have shown that biases in retrieved trace gas concentrations caused by aerosol and cloud generally are more important than those from the likely level of mis-characterisation of the instrument.

The flux inversion community has found that concentration biases adversely affect large scale estimates of surface fluxes. However, when the domain is limited, we hypothesise that biases will be less important, because they will be estimated and then eliminated during the flux inversion. As a result, reliable flux inversions might be feasible even in a polluted urban environment. This study is but one step in testing that hypothesis. Because non-random instrument errors produce biases that generally are smaller than those from aerosol, we deferred treating non-random instrument errors to future work.

We have replaced the sentence beginning at line 21 on page 14 with the following.

> The main step we have made here is a more serious treatment of the concentration retrieval errors that can be expected from a real instrument, informed by previous experience with low-earth orbiting missions OCO-2 and GOSAT. Only the random component of the instrument error is considered; non-random errors, such as those arising from imperfect radiometric calibration and imperfect characterisation of the instrument line shape function, will be considered in future work.

Minor Comments

Minor comment 1

Title : I would suggest a more appropriate title such as Potential of geostationary Geo-

Carb mission to estimate surface emissions of CO2, CH4 and CO in polluted urban environment. Case study Shanghai.

Agreed and amended as suggested.

Minor comment 2

It is not totally clear to me what the main differences are with the simulations by Polonsky et al., 2014. It seems they have done quite similar simulations.

The Polonsky et al. (2014) paper floats an idea; this paper begins the rigorous assessment. The differences are many and significant, and include the following.

1. The present paper uses WRF-Chem to model the atmosphere with all its constituents coupled. Thus, aerosol, cloud and trace gas concentrations are simulated consistently.

2. The aerosol loading over Shanghai is significantly higher in the present paper than in the simulations considered by Polonsky et al. (2014).

3. Polonsky et al. (2014) assumed a geoCARB-like instrument in polar rather than geostationary orbit. As a result, the air-mass factors were lower, which reduced the importance of aerosol and led to error estimates for trace gas concentrations that possibly were too optimistic.

4. Polonsky et al. (2014) did not attempt to estimate the amount by which observations of spectra in the geoCARB bands would reduce the prior uncertainties in the surface fluxes of CO_2, CH_4 and CO. Instead they analysed a simplified, steady-state model of the plume emitted by a power plant, and attempted to infer

the rate of emission of CO_2. The present study includes diurnal variations and complex spatial patterns of emissions of aerosols and trace gases.

Minor comment 3

p.5, l. 2 what are 'sectional size bins'?

This is the terminology used by the authors of the MOSAIC algorithm and papers. Although we could omit the word 'sectional' without loss of meaning, we prefer to keep the MOSAIC terminology.

Minor comment 4

p.6, l. 2 not unrealistic $\longrightarrow$ not unrealistic, although somewhat lower than seen by MODIS.

As explained in the text, the tail of the MODIS histogram of aerosol optical depth (AOD) extends to slightly higher values than the histogram of simulated AOD, but the most probable AOD is approximately the same for both. We see no reason to change the text.

Minor comment 5

section 3 : not explicitly mentioned which wavelength bands are used for which target molecules. Please indicate.

We have added the gases active in each band to Table 4 on page 38.

Minor comment 6

Section 5 I find it difficult to extract what all the retrieval parameters are. Please explicitly state what parameters are retrieved. Now I have to deduce that myself in some cases indirectly from the text, for example wrt meteo parameters. Is a temperature offset retrieved ? etc. Also for the aerosols. Does the standard aerosol retrieval approach mean one parameter is retrieved representing the mixing of the two types ? Is anything fitted wrt surface albedo ? (wavelength dependence ?)

There are many parameters in the retrieval, as explained (for example) by O'Dell et al. (2012); the original ACOS algorithm contained over eighty parameters. For each aerosol type, a vertical profile of extinction coefficient is retrieved in each band. The retrieval algorithm assumes that the surface albedo varies linearly across each band. Both the offset and slope of the surface albedo are retrieved in each band. We see no reason to repeat the details in this paper, as it has been documented well elsewhere.

Minor comment 7

p.10, l4 : what threshold is meant here ? What is the role of the threshold ? what other filters are set in the PPF ?

The post-processing filter is described by Polonsky et al. (2014) and earlier authors, but we accidentally omitted the reference. In response we have modified the short paragraph beginning on line 3 of page 10 as follows, adding the reference to Polonsky et al. (2014).

Thus, two options were considered, the first with standard aerosol and the second with regionally adjusted aerosol. In addition, the threshold for retrieved aerosol optical thickness in the post-processing filter (PPF) was increased from 0.1 to 0.15 for the experiment with regionally adjusted aerosol. Details of the PPF are provided by Polonsky et al. (2014).

Minor comment 8

Section 5.2 p. 8, ;. 25 what is the h2O prior profile ?

In the section titled 'Prior meteorology' on page 9, we explained that the prior meteorology was set to the truth, and gave the reasons for this choice. Therefore the prior profiles of H_2O were those calculated by WRF-chem. They varied from pixel to pixel and from one observation time to the next. There was no single prior profile of H_2O.

Minor comment 9

p.11, l. 15 ***probably*** *more important, I think that statement can easily be checked. Please do.*

The reviewer is wrong in his assumption that the statement can be checked easily, because the experimental costs in human time and computer power are considerable. This paper identifies several avenues for improvement, some numerical and others strategic. We judge the most important by a large margin to be implementation of a true flux inversion, replacing the reduction of uncertainty calculation used in the present paper. Addressing these issues, including the one raised by the reviewer, is the subject

of ongoing work.

Minor comment 10

p.13, l. 6 monotonically

We have corrected the typo.

Minor comment 11

p.13, l. 15-17, what is meant with the remark : When analising spectra ... for GeoCarb ? is this just a suggestion for future work or actually applied here ?

The text states '[a] similar approach could be adopted for geoCARB'. The use of 'could' implies that the idea is a suggestion for future work.

Minor comment 12

p. 13, l. 21 6401 flux components, is that 6400 for CO2 and 1 constant emission factor over the whole domain for CO ?

Correct. The current text on page 10 in lines 12–15 provides the explanation.

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Minor comment 13

Fig. 1 caption : Target area ... ⟶ Target area around Shanghai ...

Agreed and modified as suggested.

Minor comment 14

Fig. 3 somewhere it should be mentioned what the strong and weak band of CO2 are (see earlier comment on identification of which bands are used for which targets)

Agreed. We added the gases active in each band to Table 4. In addition, we have identified the weak and strong bands of CO_2 in the caption of Table 4. The modified caption is as follows.

> Spectral ranges and resolutions of the geoCARB bands for the baseline configuration. The bands at 1.606 μm and 2.065 μm are referred to in the text as the 'weak' and 'strong' CO_2 bands.

Minor comment 15

Fig. 7-10 caption : The PPF .. light grey ⟶ The light grey histograms are with PPF disabled, while the coloured histograms are obtained after applying PPF

We see no advantage in the wording offered by the reviewer.

Minor comment 16

Fig. 11 caption : actual errors (retrieved ...) ⟶ actual errors (points with error bars, retrieved ..)

Agreed and modified as suggested.

Minor comment 17

Table 1 caption : Assignment of EDGAR ⟶ assignment (in fraction) of EDGAR

Agreed and modified to the following caption.

> Fractional assignment of EDGAR particulates to Aitken, accumulation and coarse modes.

Minor comment 18

Table 7 : should be made clear in the caption what disabled en enabled means below Cloud

We have added the following short paragraph before the paragraph starting at line 10 on page 11.

> Three experiments were conducted, as indicated in the first three columns of Table 7. In experiment 1, the aerosol over Shanghai was simulated, but the optical thicknesses of water and ice clouds were set to zero. The retrieval algorithm used the ACOS aerosol scheme with water cloud, ice cloud

and two types of aerosol. The retrieval algorithm did not assume that the atmosphere was cloud free; it retrieved optical thickness profiles of aerosol, cloud water and cloud ice, layer by layer. Experiment 2 was similar, except that the radiance simulations of the forward model included the contributions of cloud water and cloud ice. Lastly, the forward simulations for experiment 3 included both aerosol and cloud, but the retrieval algorithm used adjusted optical properties for aerosol as described under the sub-section 'Prior aerosol' above.

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