Comment on *The ICAD (Iterative Cavity Enhanced DOAS) Method* by Martin Horbanski et al.

This paper is the third in row with the publications of Platt et al., 2009 and Meinen et al, 2010. It describes the experimental setup of an improved broadband (BB) Cavity-Enhanced (CE) DOAS instrument which uses the well-known DOAS evaluation principle for the retrieval of trace gas concentrations from absorption spectra.
While in 'conventional' DOAS spectroscopy the optical path length between light source and detector is constant and precisely known this is not the case if an optical cavity is used to create the long absorption path. In this paper the authors thoroughly discuss the aspect of the shortening of the photon lifetime in the CEAS cavity due to the presence of absorbing trace species which leads to non-linear changes of the 'average optical path length' as a function of the trace gas concentration. 'Traditional' broad-band CEAS evaluation schemes (see papers of Fiedler et al.) inherently account for these path changes by measuring the absolute total light intensity. Hence these instruments require long-term stable light sources with very small intensity variations and mechanically rigid optical setups in order to assure equal light intensities at the detector during the measurements of the cavity transmission without and with absorbers.
In this paper the authors present a new spectral retrieval approach to overcome the above mentioned technically challenging restrictions. An iterative algorithm (ICAD CE-DOAS) was developed which, on the one hand, models the optical path length reduction as a function of the calculated actual optical density (OD) in the cavity. On the other hand the iterative method introduces the important DOAS advantage of being independent on the absolute light intensity into the 'CEAS world', however, at the expense of a significantly more complex data analysis.
The paper is certainly appropriate for publication in AMT after the comments below are appropriately answered.

Page 2, lines 18-30: I disagree with the statement 'the so far presented instruments can only be operated in a laboratory or at least in laboratory similar condition'. CEAS instruments already have been operated under harsh field measurement conditions so this statement needs rephrasing.

Page 3, lines 7-10: Mentioning of 'a predecessor CE-DOAS instrument ...' should be removed from the text because it does not provide any useful information to the topic of the manuscript.

Page 4, lines 26-29: I understand that the authors like to express how 'tolerant' their instrument is to intensity fluctuations but since the strong temperature dependency of the LED output power and spectrum is very well known in the community, nearly all LED driven CEAS instruments use temperature stabilization, usually by accurate and cheap Peltier cooling. Hence I think this is not a generally valid argument.

Sects. 2.1 and 2.2, Eqs. 2, 3, and 4 need some clarification to avoid confusion. The molecular absorption cross sections used in Eqs. 2 and 3 are the total absorption cross section, i.e., sum of differential part $\sigma'(\lambda)$ plus broadband part, called $\sigma_0(\lambda)$ in Sect. 2.2.
In contrast Sect. 2.2, Eq. 4 the DOAS approach just considers the differential part of the total cross section. Consequently the differential optical density should be denoted $D'_{model}(\lambda)$.

Page 8, lines 1 ff: The authors state that for cases when no particle filters in the inlet are acceptable due to trace gas losses on the surface the aerosol extinction term $\varepsilon_{aerosol}(m,\lambda)$ (Eq. 8) must be estimated 'by a suitable parametrization'.
However in Fig. 4 (first equation in the box) the broadband aerosol extinction ε_b (see Eq.5) or $\varepsilon_{aerosol}(m,\lambda)$ (see Eq. 8), respectively, is omitted. The authors justify this by simply stating (page 9, lines 1-3) that the present paper only regards the aerosol FREE case and that 'in the presence of aerosols the removal of broadband absorption, as part of the DOAS analysis, needs to be performed with care not to interfere with the aerosol absorption.'

- Please explain what is meant by ‘suitable parametrization'.
- Could it be that in the case of strong aerosol extinction the iterative technique does not necessarily lead to a bijective solution?

I think this aspect must be discussed in greater detail in this paper in order to help interested future users of the ICAD technique to understand its potential limitations.

Page 8, lines 7: What is meant by 'if all absorbers have sufficiently wavelength dependent absorption structures'? I assume this only refers to the differential part of the trace gas absorption spectra. How does this help to overcome the broadband aerosol absorption?

Page 9, Fig.5: This figure shows the iterative procedure to calculate the concentration from theoretical CEAS spectra for an extremely high NO_2 concentration.
In principle the figure is well suited for this topic, however due to the exclusion of the fundamental shot noise the archived minimal peak-to-peak noise levels of the residual (Fig. 5d) of 10^{-9} to 10^{-6} might give the reader false expectations what can be achievable with ICAD. It should be made clear that in reality the min. expectable noise levels are fundamentally limited by the photon shot noise.

Page 12, Sect. 3.1.2: In Fig. 6 it is shown that the retrieval of trace gas concentrations by the BB-CEAS method linearly depends on the change of radiation intensity between zero-air and ambient air measurements. The simulations were done up to 90% intensity differences.
However the dramatic results (of 90% intensity change !) shown here should be put into perspective to realistically achievable intensity fluctuations of presently used temperature stabilized light sources and CEAS cavities, which I assume to be less than a percent.
Compare the following discussion of the requirements of equals light intensities during the calibration of the path length in the air filled cavity on page 15, line 17 where a relative intensity drift of $5x10^{-4}$ over some minutes was denoted.

Page 15, Section 4.3.1: It is the measurement PRECISION, not accuracy !

Figures:

Fig. 2: Shows the light path length as function of wavelength.
There are three extinction processes contributing to the path length which should be explained to the reader for clearness.
Firstly, on the second ordinate the absolute contribution of mirror transmission losses should be indicated.
Secondly, the absolute contribution of Rayleigh scattering in synthetic air over the spectral interval should be quantified.
Thirdly, the loss due to absorption of the water dimer should be shown in relation to both other losses.

Fig. 5 should be moved between Sect. 3.1 and 3.1.1

Fig. 8: The text should read: 'The bottom graph shows the time series OF THE 1-σ ERROR OF THE NO_2 CONCENTRATION for different integration times'

Typos, word replications, etc.:

p4 line 22 / p7 line 12 / p5, line 4 / p7, line 12 / p8 / in Figure 4: ‘Rayleigh extinction’
p8, line 8 / p9, caption to Fig.5 / p11, line 3 / p20, line 1