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Sensitivity studies of the air mass factors (AMF) used to retrieve glyoxal Vertical Column Densities from space

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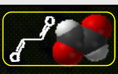
ABSTRACT

Glyoxal (CHO.CHO), the smallest dicarbonyl organic compound, is known to be a key intermediate product of the oxidation of VOCs emitted from anthropogenic activities, biogenic processes and biomass burning. Recently, glyoxal has been detected in atmospheric aerosols and has been suggested to form a significant part of the missing secondary organic aerosols, which is not reproduced by the models. Monitoring CHO.CHO from space provides, therefore, useful information concerning a) the VOC oxidation and b) the secondary organic aerosol formation (SOA) that are key processes of atmospheric pollution.

The determination of the amounts of trace gasses from space is based on several a priori assumptions used in the computation of an air-mass factor (AMF). The latter is used for the transformation of the Slant Column Densities to Vertical Column Densities (VCDs). The AMF depend on the radiative transfer properties of the atmosphere. Parameters such as: the *surface spectral reflectance* of a ground scene, the *surface pressure*, the *viewing geometry*, the *clouds*, the assumption for the *aerosol vertical distribution* and its *optical thickness*, the *solar zenith angle* and finally, the assumed *vertical distribution of* CHO.CHO in the lowermost troposphere, are determining the AMFs. This work presents some of the key uncertainties in the AMF calculations related to the boundary layer height, aerosol layer height, aerosol optical depth, albedo and single scattering albedo.

Species of interest: CHO.CHO - Sources and Sinks

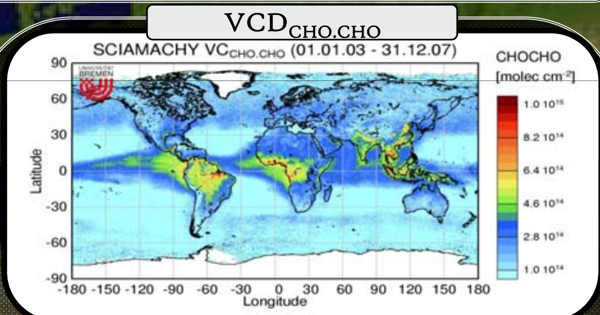
CHOCHO is formed by the oxidation of NMHC. Contrary to HCHO no direct sources are expected. This makes CHOCHO a good indicator of the VOC oxidation.



Glyoxal

The main known sinks of CHOCHO are: a) the reaction with the OH radicals and the b) photolysis leading to an estimated lifetime of 2h.
c) Another sink, although still highly uncertain, is the reversible, or irreversible uptake of CHO.CHO on/in aerosols and clouds.

$VCD_{CHO.CHO}$



As highlighted at the abstract, CHO.CHO is expected to give insights into the homogeneous and multiphase/ heterogeneous atmospheric processes driven by the VOC oxidation. Satellite remote sensing can provide a variety of useful data for this type of research as it is the means to calculate the glyoxal vertical column densities on a global scale. The multiannual (2003 –2007) composite map of $VCD_{CHO.CHO}$ shows that South America, Africa, India, Indonesia and Asia (mainly South-eastern China) are among the dominant regions where high values of CHOCHO ($\geq 6\cdot 10^{14}$ molec·cm^{-2}) are retrieved. At higher latitudes, moderate values of $VCD_{CHO.CHO}$ of about $3\cdot 10^{14}$ molec·cm^{-2} are discernible, for example above North America ground scenes, Europe and Australia. Notably, high column amounts of CHO.CHO are also observed above water suggesting enhanced biogenic activity. Due to the short lifetime of CHOCHO of about 2-3 hours, these high values are expected to originate from the region sources of the precursor VOCs. Indeed, depending on the season, these regions are characterized by strong biogenic emissions, biomass burning and pollution induced from anthropogenic activities.

To convert the SCD_{CHOCHO} (retrieved by the DOAS technique), to the VCD_{CHOCHO} column densities presented above, the monthly mean 2D-AMF calculated by Wittrock, (2006) were used. However, in order to improve the accuracy of the results it is essential to understand the key uncertainties linked to the AMF calculation.

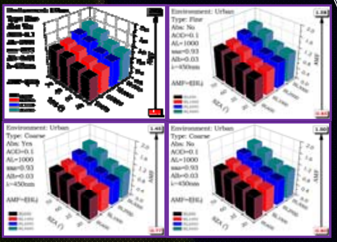
Motivation

Improving the air mass factors used to retrieve the $VCD_{CHO.CHO}$ by studying their key uncertainties

The task of the current study is to present some of the uncertainties related to the AMF calculations (with the emphasis given to aerosol properties). In specific the behavior of the AMF as a function of the boundary layer (BL) variation, the aerosol layer (AL) height, the aerosol optical depth (AOD), the albedo and the single scatteting albedo (ssa) is depicted below.

① AMF = f (BL)

For this test we used a homogeneous aerosol layer height well mixed into different boundary layer heights (see graph below). The AMF increases when the BL increases. Higher SZA corresponds to higher AMF. Coarse and absorbing aerosols show lower AMF values in comparison to fine and non absorbing ones, respectively.

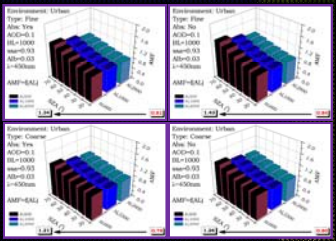


+3%

$$AMF_{BL600} < AMF_{BL1000} < AMF_{BL2000} < AMF_{BL3000}$$

+17%

② AMF = f (AL)



In this case the variable is the aerosol layer which is homogeneously mixed with the trace gasses into a constant BL (1km). For the case of AL=2km we assume that the first 1km is well mixed into BL.

-9%

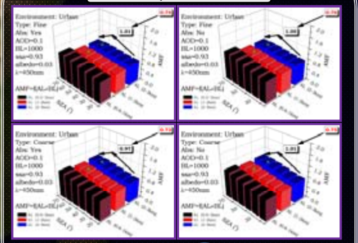
$$AMF_{AL600} > AMF_{AL1000} > AMF_{AL2000}$$

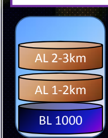
-16%

As the aerosol layer height increases the AMF values decrease as a result of the shielding to the CHO.CHO absorption signal.

③ AMF = f (AL≠BL)

Case study 1: (AOD = 0.1)





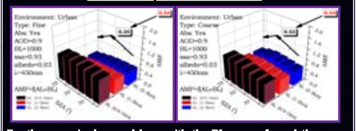
Similarly to ② the variable is again the aerosol layer. The main difference is that the AL is not mixed with the BL but lays above it. The shielding to the CHO.CHO absorption signal is greater. The computed AMF for the $AL_{1\text{-}2\,km}$ and the $AL_{2\text{-}3\,km}$ are about the same.

-10%

$$AMF_{AL:0.6\text{-}1km} > AMF_{AL:1\text{-}2km} \approx AMF_{AL:2\text{-}3km}$$

-9%

Case study 2: (AOD = 0.9)



For the non-mixed aerosol layer with the BL, we performed the same sensitivity test as before but with an increased aerosol load of 0.9 . As a result the aerosol layer produces a massive shielding. A vast reduction in the computed AMF is now observed.

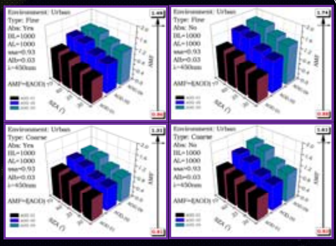
-47%

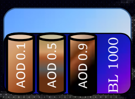
$$AMF_{AL:0.6\text{-}1km} > AMF_{AL:1\text{-}2km} > AMF_{AL:2\text{-}3km}$$

-52%

④ AMF = f (AOD)

This sensitivity case study analyzes the AMF changes as a function of the Aerosol Optical Depth or in other words the degree of which aerosols prevent the transmission of light.
It was found that when the AOD increases the AMF values are higher for the case that the AL and the BL are well mixed inside the 1km height. The results vary according to the type of aerosol (fine /coarse, absorbing /non-absorbing)



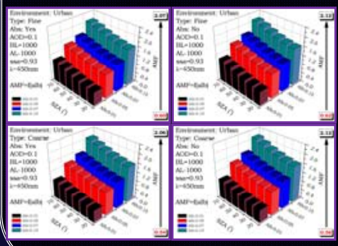


+27%

$$AMF_{AOD0.1} < AMF_{AOD0.5} < AMF_{AOD0.9}$$

+39%

⑤ AMF = f (albedo)

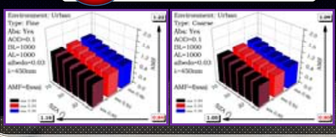


It was found that the most crucial parameter affecting AMF values is the proportion of the radiation reflected at a surface (albedo). High reflectivity increases drastically the AMF values.

$$AMF_{alb0.01} < AMF_{alb0.05} < AMF_{alb0.07} < AMF_{alb0.15}$$

+64%
+88%
+146%

⑥ AMF = f (ssa)



The last test involves the calculations of the AMF as a function of the single scattering albedo (ssa). It was found that when $0.89<ssa<0.96$ the AMF encounters only small changes (up to 4%).

SCIATRAN Parameterization

Radiative tranfer model: SCIATRAN (the yellow color denotes the basic run parameters)
Wavelengths: 425, 437.5, 440, 450nm (main CHO.CHO absorption band at 457nm).
SZA: 20°, 30°, 40°, 50°, 60°, 70°
Aerosol Optical Depth (AOD) → 0.1, 0.5, 0.9
Aerosol height (AL) → 0.6km, 1km, 2km
Albedo → 0.01, 0.03, 0.05, 0.07, 0.15
Absorbing → Yes/No
Boundary Layer Height (BL)→0.6km, 1km, 2km
Single scattering albedo (SSA) → 0.89, 0.93, 0.96
Size → Fine or coarse
Environment → Urban

ACKNOWLEDGMENTS

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Conclusions

- ✓ According to the sensitivity study performed with the radiative transfer model SCIATRAN, the most important parameter affecting the computed AMFs is the albedo. When the surface reflectivity increases from 0.01 to 0.07 , the AMF values increase up to about 90%. ⑤
- ✓ When the aerosol layer is homogeneously mixed with the trace gas into the BL and ,
  - a) the BL elevates, the AMF increases due to the increased sensitivity as more photons reach the detector; ①
  - b) the AOD increases, the AMF also increases. The effect is larger for the non-absorbing aerosols due to the increased scattering; ④
  - c) the BL remains constant (1km) but the AL raises (from 0.6km to 2km), the AMF decreases due to the shielding of the trace gas signal. ②
- ✓ When the aerosol layer lays above the BL (and not mixed with it), the shielding effect leads to lower AMFs. The shielding is even larger when the AOD increases. ③
- ✓ In general coarse and/or absorbing aerosols lead to lower AMFs in comparison to fine and non absorbing ones respectively.