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## Acetylene (C<sub>2</sub>H<sub>2</sub>) and hydrogen cyanide (HCN) from IASI satellite observations: global distribution, validation, and comparison with model.

Valentin Duflot, Catherine Wespes, Lieven Clarisse, Daniel Hurtmans, Yasmine Ngadi, Nicolas Jones, Claire Paton-Walsh, Juliette Hadji-Lazaro, Corinne Vigouroux, Martine de Mazière, et al.

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Global  
measurements of  
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V. Dufлот et al.

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# Acetylene (C<sub>2</sub>H<sub>2</sub>) and hydrogen cyanide (HCN) from IASI satellite observations: global distributions, validation, and comparison with model

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

We present global distributions of  $C_2H_2$  and HCN total columns derived from the Infrared Atmospheric Sounding Interferometer (IASI). These distributions are obtained with a fast method allowing to retrieve  $C_2H_2$  abundance globally with a 5% precision and HCN abundance in the tropical (subtropical) belt with a 10% (30%) precision. IASI data are compared for validation purposes with ground-based Fourier Transform Infrared (FTIR) spectrometer measurements at four selected stations. We show that there is an overall agreement between the ground-based and space measurements. Global  $C_2H_2$  and subtropical HCN abundances retrieved from IASI spectra show the expected seasonality linked to variations in the anthropogenic emissions and seasonal biomass burning activity, as well as exceptional events, and are in good agreement with previous spaceborne studies. IASI measurements are also compared to the distributions from the Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4). Seasonal cycles observed from satellite data are reasonably well reproduced by the model. However, the model seems to overestimate (underestimate) anthropogenic (biomass burning) emissions and a negative global mean bias of 1% (16%) of the model relative to the satellite observations was found for  $C_2H_2$  (HCN).

## 1 Introduction

Hydrogen cyanide (HCN) and acetylene (or ethyne,  $C_2H_2$ ) are ubiquitous atmospheric trace gases with medium lifetime, which are frequently used as indicators of combustion sources and as tracers for atmospheric transport and chemistry. For HCN, biomass burning is the primary source, followed by fossil fuel combustion and higher plants, bacteria and fungi (Cicerone and Zellner, 1983; Li et al., 2000), and its primary sink is thought to be ocean uptake (Li et al., 2000). For  $C_2H_2$ , biofuel combustion is considered as the dominant source, followed by fossil fuel combustion and biomass burning

ACPD

15, 14357–14401, 2015

### Global measurements of HCN and $C_2H_2$ from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



*Atmosphit* (Coheur et al., 2005). In the cases of HCN and C<sub>2</sub>H<sub>2</sub>, the accuracy of the retrievals has been recently improved by taking into consideration the CO<sub>2</sub> line mixing in the radiative transfer model (Dufлот et al., 2013). This retrieval method, relying on spectral fitting, needs a high computational power and is time consuming, especially when a large number of spectra has to be analyzed and fitted. This is therefore not suitable for providing global scale concentrations distributions of these trace gases in a reasonable time.

One of the commonly used methods for the fast detection of trace gases is the brightness temperature difference (BTD) between a small number of channels, some being sensitive to the target species, some being not. Such a method has been used from IASI spectra for sulfur dioxide (SO<sub>2</sub>) (Clarisse et al., 2008) and ammonia (NH<sub>3</sub>) (Clarisse et al., 2009). It is of particular interest in operational applications (quick alerts) or when large amounts of data need to be processed. However, relying on a cautious selection of channels to avoid the contamination with other trace gases, the BTD method does not fully exploit all the information contained in hyperspectral measurements. Especially, low concentrations of the target species may not be detected with such a method.

Walker et al. (2011) presented a fast and reliable method for the detection of atmospheric trace gases that fully exploits the spectral range and spectral resolution of hyperspectral instruments in a single retrieval step. They used it to retrieve SO<sub>2</sub> total column from a volcanic plume and NH<sub>3</sub> total column above India. More recently, Van Damme et al. (2014) presented a retrieval scheme to retrieve NH<sub>3</sub> from IASI spectra based on the work of Walker et al. (2011), and introduced a metric called Hyperspectral Range Index (HRI). We use in the present study a similar approach.

### 2.2.1 Hyperspectral Range Index (HRI)

The method used in this study is a non-iterative pseudo retrieval method of a single physical variable or target species  $x$  expressed as, following the formalism developed

by Rodgers (2000):

$$\hat{x} = x_0 + (K^T S_e^{\text{tot}-1} K)^{-1} K^T S_e^{\text{tot}-1} (y - F(x_0)) \quad (1)$$

where  $y$  is the spectral measurements,  $x_0$  is the linearization point,  $F$  is the forward model (FM),  $S_e^{\text{tot}}$  is the covariance of the total error (random + systematic), and the Jacobian  $K$  is the derivative of the FM to the target species in a fixed atmosphere.

$S_e^{\text{tot}}$  can be estimated considering an appropriate ensemble of  $N$  measured spectra which can be used to build up the total measurement error covariance  $S_y^{\text{obs}}$ :

$$S_e^{\text{tot}} \simeq \frac{1}{N-1} \sum_{j=1}^N (y_j - \bar{y})(y_j - \bar{y})^T = S_y^{\text{obs}} \quad (2)$$

where  $\bar{y}$  is the calculated mean spectrum for the ensemble.

To generate  $S_y^{\text{obs}}$ , we randomly chose 1 million spectra observed by IASI all over the world, above both land and sea, during the year 2009. Then, we applied a BTD test to remove the spectra contaminated by the target species. For HCN ( $\text{C}_2\text{H}_2$ ), the wavenumbers 716.5 and 732  $\text{cm}^{-1}$  (712.25 and 737.75  $\text{cm}^{-1}$ ) were used as reference channels and 712.5  $\text{cm}^{-1}$  (730  $\text{cm}^{-1}$ ) was used as test channel (Fig. 1, middle panel).

Given the medium lifetimes of the target species (few weeks for  $\text{C}_2\text{H}_2$  to few months for HCN), and the limited accuracy of the BTD test due to the weak spectral signatures of the target species, it is likely that such randomly chosen and filtered spectra still contain a small amount of the target species whose signal may come out from the noise. This limitation decreases the sensitivity of the method, which is discussed in Sect. 2.2.3.

The spectral ranges considered to compute the  $S_y^{\text{obs}}$  matrices are 645–800  $\text{cm}^{-1}$  for HCN and 645–845  $\text{cm}^{-1}$  for  $\text{C}_2\text{H}_2$  (Fig. 1, top panel). These ranges were chosen as they include parts of the spectrum which have a relatively strong signal from the target species but also from the main interfering species ( $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and  $\text{O}_3$ , Fig. 1, bottom panel) in order to maximize the contrast with the spectral background.

Global measurements of HCN and  $\text{C}_2\text{H}_2$  from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



in Sect. 2.2.3). Each of the constructed profile has been associated with a spectrum through the FM of *Atmosphit* considering standard absorption profiles. The associated values of  $\text{HRI}_{\text{HCN}}$  and  $\text{HRI}_{\text{C}_2\text{H}_2}$  have then been computed for each of the simulated spectra. Figure 2 shows the look up tables (LUTs) of  $\text{HRI}_{\text{HCN}}$  (top) and  $\text{HRI}_{\text{C}_2\text{H}_2}$  (bottom) as a function of the abundance of the target molecule and of the altitude of the polluted layer in a standard tropical modeled atmosphere (Anderson et al., 1986). Similar LUTs have been computed for standard temperate (US standard atmosphere) and polar (Anderson et al., 1986) atmospheres (data not shown). The satellite viewing angles were taken into account in the HRI calculation similarly to Van Damme et al. (2014). One can see that, for a given atmosphere and for a given altitude of the polluted layer, the abundances of both species linearly depend on the HRI value, which validates Eq. (5). For a given atmosphere atm and a given species  $X$ , the different values of  $B$  with respect to the altitude  $z$  of the polluted layer will be noted  $b_{X_{\text{atm}}}(z)$  and  $k_{X_{\text{atm}}}(z)$  in the following.

Figure 3 shows the normalised Jacobians of the FM for HCN and C<sub>2</sub>H<sub>2</sub> averaged over the spectral ranges given in Sect. 2.2.1 (645–800 cm<sup>-1</sup> for HCN and 645–845 cm<sup>-1</sup> for C<sub>2</sub>H<sub>2</sub>) and for each of the three standard modeled atmospheres. These Jacobians express the sensitivity of the FM, i.e. both the radiative transfer model and IASI (through its instrumental function), to the target species abundance  $X$  in a fixed atmosphere atm:

$$K_{X_{\text{atm}}} = \left[ \frac{\partial F_{\text{atm}}}{\partial X}(z_1) \dots \frac{\partial F_{\text{atm}}}{\partial X}(z_n) \right] = \left[ k_{X_{\text{atm}}}(z_1) \dots k_{X_{\text{atm}}}(z_n) \right] \quad (6)$$

We then obtain the coefficients  $B_{X_{\text{atm}}}$  by multiplying the  $b_{X_{\text{atm}}}(z)$  by the value of the Jacobian at the altitude  $z$ :

$$B_{X_{\text{atm}}} = \sum_{i=1}^n \left( b_{X_{\text{atm}}}(z_i) \times k_{X_{\text{atm}}}(z_i) \right) \quad \text{with} \quad \sum_{i=1}^n k_{X_{\text{atm}}}(z_i) = 1 \quad (7)$$

Applying this method to the three standard modeled atmospheres (tropical, temperate and polar), we get a  $B_X$  value for each, which we have associated with the corresponding range of latitude ( $[\pm 20^\circ]$ ,  $[\pm 45^\circ : \pm 60^\circ]$ ,  $[\pm 75^\circ : \pm 90^\circ]$ , respectively), and linearly interpolated between. Figure 4 gives the resulting values of  $B_{\text{HCN}}$  (blue) and  $B_{\text{C}_2\text{H}_2}$  (green) in function of the latitude.

### 2.2.3 Sensitivity and stability of the method

The sensitivity of the method can be assessed from the Jacobians presented in Fig. 3. For HCN, one can see that there is no sensitivity at the surface and above  $\sim 30$  km, and the altitude of the sensitivity peak is located close to the tropopause at  $\sim 9$ ,  $\sim 11$  and  $\sim 14$  km for the polar, temperate and tropical atmospheres, respectively. For  $\text{C}_2\text{H}_2$ , there is no sensitivity above  $\sim 20$  km, and the maximum sensitivity is reached at  $\sim 8$ ,  $\sim 10$  and  $\sim 11$  km for the polar, temperate and tropical atmospheres, respectively.

The HRIs presented here above are sensitive to the abundance of the target species – this is what they are made for – and to their vertical distribution. However, the measured column amount may also depend on: (1) the proper suppression of the spectral background, (2) the conditions of thermal contrast with the surface (TC), and (3) the accuracy of the FM to simulate the spectra used to build up the LUTs. The latter was discussed already by Dufлот et al. (2013). In order to test the impact of the two first factors (spectral background suppression and TC) on the retrieved column amount, HCN and  $\text{C}_2\text{H}_2$  profiles have been constructed with varying TC and concentrations of the interfering and target species. The TC is defined here as the difference between the skin (surface) temperature and that of the air at an altitude of 1.5 km. These variations in interfering species abundances and TC were considered to be independent and were taken within the range  $\pm 2\%$  for  $\text{CO}_2$  and  $\pm 20\%$  for  $\text{H}_2\text{O}$  and  $\text{O}_3$ , and in the range  $\pm 10\text{K}$  for the TC. For a fixed column amount of the target species, the HRIs were compared one by one to a HRI corresponding to a standard spectrum (i.e. with background concentrations of the interfering species and a TC equal to zero) and if the

## Global measurements of HCN and $\text{C}_2\text{H}_2$ from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion









## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



sonality and interannual variability matches reasonably well that of the ground-based measurements (correlation coefficient of 0.40 for the entire daily mean dataset, and of 0.72 for the monthly mean data set) but with the IASI columns being biased high by  $1.10 \times 10^{15}$  molec cm<sup>-2</sup> (107%). Such a high bias between the two datasets could be due to the difference between space and ground-based instruments sensitivity (Figs. 3 and 7). One can also notice that the C<sub>2</sub>H<sub>2</sub> and HCN peaks are higher in 2010. As South American biomass burning plumes are known to impact trace gases abundance above Reunion Island (Edwards et al., 2006a, b; Dufлот et al., 2010), these 2010 higher peaks are probably due to the 2010 great Amazonian fires (Lewis et al., 2011) influence.

At Wollongong HCN peaks also in October–November due to the Southern Hemisphere biomass burning season (Paton-Walsh et al., 2010). We find maxima of around  $11 \times 10^{15}$  molec cm<sup>-2</sup> in October 2010, which is, similarly to Reunion Island, very likely to be a signature of the great Amazonian fires as South American biomass burning plumes are known to impact trace gases abundance above Australia (Edwards et al., 2006a, b). The seasonality and interannual variability matches well that of the ground-based FTIR measurements (correlation coefficient of 0.55 for the entire daily mean dataset, and of 0.83 for the monthly mean data set), with the IASI columns being biased low by  $0.48 \times 10^{15}$  molec cm<sup>-2</sup> (10%).

At Izaña HCN peaks in May–July due to the biomass burning activity occurring in Northern America and Europe (Sancho et al., 1992). We find maxima of around  $8 \times 10^{15}$  molec cm<sup>-2</sup>. The seasonality and interannual variability matches poorly that of the ground-based FTIR measurements for HCN (correlation coefficient of 0.28 for the entire daily mean dataset, and of 0.64 for the monthly mean data set), with the IASI columns being biased high by  $0.45 \times 10^{15}$  molec cm<sup>-2</sup> (11%). One can notice that HCN total columns as measured by ground-based FTIR are below the HCN stability threshold in boreal winter, which may result in erroneous IASI measurements (because unstable) and explain this poor match between the two datasets.

For C<sub>2</sub>H<sub>2</sub> at the Jungfrauoch site, the agreement between IASI and the ground-based retrieved columns is good (correlation coefficient of 0.70 for the entire daily mean

dataset, and of 0.85 for the monthly mean data set), with the IASI columns being biased low by  $0.15 \times 10^{15} \text{ molec cm}^{-2}$  (12%), opposite to the observations at Reunion. The larger columns observed in late winter are caused by the increased  $\text{C}_2\text{H}_2$  lifetime in that season (caused by the seasonal change in OH abundance) (Zander et al., 1991), and we find corresponding maxima of up to  $4 \times 10^{15} \text{ molec cm}^{-2}$ .

### 3.2 IASI Global distributions

We focus in this section on the description of the  $\text{C}_2\text{H}_2$  and HCN distributions retrieved from IASI spectra. For practical reasons, the figures used in this section also show simulated distributions that will be analyzed afterwards.

The left panels of Figs. 9 and 10 provide the seasonal global and subtropical distributions of  $\text{C}_2\text{H}_2$  and HCN total columns, respectively, as measured by IASI and averaged over the years 2008 to 2010.

Looking at IASI measurements (Figs. 9 and 10 – left panels), one can notice the following main persisting features for both  $\text{C}_2\text{H}_2$  and HCN:

- the hot spots mainly due to the biomass burning activity occurring in Africa and moving southward along the year (Sauvage et al., 2005; van der Werf et al., 2006);
- the hot spot located in South East Asia being likely a combination of biomass burning and anthropogenic activities;
- the transatlantic transport pathway linking the African west coast to the South American east coast and moving southward along the year (Edwards et al., 2003, 2006a, b; Glatthor et al., 2015).

The following seasonal features can also be observed:

- the transpacific transport pathway linking Eastern Asia to Western North America, especially in March-April-May (MAM) (Yienger et al., 2000);



## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- the transport pathway from Southern Africa to Australia in June-July-August (JJA) and September-October-November (SON) (Annegarn et al., 2002; Edwards et al., 2006a, b);
- the transport pathway linking South America (especially Amazonia) to Southern Africa and Australia during the SON period (Edwards et al., 2006a, b; Glatthor et al., 2015);
- the transport of the northern African plume over southern Asia to as far as the eastern Pacific by the northern subtropical jet during the MAM period (Glatthor et al., 2015);
- the Asian monsoon anticyclone (AMA), which is the dominant circulation feature in the Indian-Asian upper troposphere-lower stratosphere (UTLS) region during the Asian summer monsoon, spanning South East Asia to the Middle East and flanked by the equatorial and sub-tropical jets (Hoskins and Rodwell, 1995). The AMA is a known region of persistent enhanced pollution in the upper troposphere, linked to rapid vertical transport of surface air from Asia, India, and Indonesia in deep convection, and confinement by the strong anticyclonic circulation (Randel et al., 2010). The enhanced abundance of C<sub>2</sub>H<sub>2</sub> and HCN within the AMA in JJA observed by IASI is in accordance with previous studies (Park et al., 2008; Randel et al., 2010; Parker et al., 2011; Glatthor et al., 2015); however, one should keep in mind that this enhanced abundance measured by IASI is likely due to the combination of this pollution uplift and confinement with the higher sensitivity of the method in the upper troposphere (Fig. 2).

One can also notice the very good agreement between the seasonal HCN distributions shown in our Fig. 10 and the ones published recently in Glatthor et al. (2015, Fig. 3).

Figures 11 and 12 show the C<sub>2</sub>H<sub>2</sub> and HCN total columns time series, respectively, as measured by IASI (red dots) with the associated SD (light red lines) for each of the zones defined in Fig. 6.

**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



In Northern America, Europe and Boreal Central Asia (Fig. 11 – Zones NAM, EUR and BCA), C<sub>2</sub>H<sub>2</sub> peaks in late boreal winter due to the increased C<sub>2</sub>H<sub>2</sub> lifetime as already noticed over Jungfrauoch (Fig. 8). The boreal summer 2008 California wildfires event (Gyawali et al., 2009) is clearly visible in the NAM plot, as well as the August 2009 Russian wildfires in the NAM, EUR and BCA plots (Parrington et al., 2012; R'honi et al., 2013).

In North Central America (Fig. 12 – Zone NCA), the annual HCN peak in April–June is driven by local fire activity (van der Werf et al., 2010).

In South America, Southern Africa and Australia (Figs. 11 and 12 – Zones SAM, SAF and AUS), the Southern Hemisphere biomass burning season clearly drives the C<sub>2</sub>H<sub>2</sub> and HCN peaks in September–November each year. The signature of the great 2010 Amazonian fires (Lewis et al., 2011) is visible on each of the these three Zones, South American fire plumes being known to impact Southern Africa and Australia (Edwards et al., 2003, 2006a, b). The February 2009 Australian bush fires (Glatthor et al., 2013) are also noticeable on Zone AUS for both species.

In Northern Africa (Figs. 11 and 12 – Zone NAF), C<sub>2</sub>H<sub>2</sub> and HCN peak in boreal winter because of the biomass burning activity occurring in the Zone, and peak also in boreal summer because of the European and South Mediterranean fires (Van der Werf et al., 2010).

In South East Asia (Figs. 11 and 12 – Zone SEA), the observed C<sub>2</sub>H<sub>2</sub> and HCN peaks in July–September and January–March are due to local fire activity (Fortems-Cheiney et al., 2011; Magi et al., 2012). Additionally, the July–September peaks are also likely due to the combination of the pollution uplift and confinement within the AMA with the higher sensitivity of the method in the upper troposphere.

In Equatorial Asia (Figs. 11 and 12 – Zone EQA), local fire activity is visible in July–October, as well as the South East Asian fire activity in January–March (Fortems-Cheiney et al., 2011; Magi et al., 2012). The high biomass burning activity occurring in Indonesia from July to December 2009 (Yulianti et al., 2013; Hyer et al., 2013) is also clearly noticeable.







In Table 2, for  $C_2H_2$ , the correlation coefficients are good ( $\geq 0.6$ ) to very good ( $\geq 0.9$ ) except for the zones SAM (South America), SEA (South East Asia) and EQA (Equatorial Asia). For HCN, the correlation coefficients are good ( $\geq 0.6$ ) except for the zones NCA (North Central America), NAF (Northern Africa), SEA and EQA.

5 For South America (Zone SAM), correlation coefficient is not as good for  $C_2H_2$  ( $R = 0.54$ ) due to a backward shift of the species abundance peaks in years 2008 and 2009: in the model, this increase occurs from July to October while observations (and previous studies, e.g. van der Werf et al., 2010) show an increase from August to December. This backward shift is also visible for HCN (Fig. 12), but to a lesser extent.

10 For South East Asia and Equatorial Asia (Zones SEA and EQA), the low correlation coefficients (cf. Table 2) can be attributed to the difficulty of locating precisely with the model the intercontinental convergence zone (ITCZ) which drives the long-range transport of  $C_2H_2$  and HCN-loaded plumes into the zone. Additionally, for Equatorial Asia, the too low fire emissions considered in the model for Indonesia from July to December 2009 may also be a cause for these low correlation coefficients.

15 For HCN in northern Africa (Zone NAF), correlation coefficient is very low ( $R = 0.07$ ) because the model sets the abundance peaks around August while observations show peaks occurring around December, which is in accordance with previous studies (van der Werf et al., 2010). This inadequate timing for HCN in the model simulations could be due to an overestimation of the Southern African contribution to the Northern African loading and is visible on Fig. 10 (JJA).

## 4 Conclusions

25 We have presented a fast method to retrieve HCN and  $C_2H_2$  total columns from IASI spectra. The sensitivity of this method to the two species is mostly in the mid-upper troposphere. With this method,  $C_2H_2$  total columns can be retrieved globally with 5% precision, while HCN abundances can be retrieved for abundances greater than

Global  
measurements of  
HCN and  $C_2H_2$  from  
IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



$0.28 \times 10^{16} \text{ molec cm}^{-2}$  with 10 % precision in the  $\pm 20^\circ$  latitudinal band and with 30 % precision in the  $[\pm 35^\circ : \pm 20^\circ]$  latitudinal band.

Total columns have been retrieved globally for a three year period and compared to routine FTIR measurements available at Reunion Island (HCN and  $\text{C}_2\text{H}_2$ ), Wollongong (HCN), Jungfraujoch ( $\text{C}_2\text{H}_2$ ), and Izaña (HCN). The comparison between IASI and FTIR retrieved total columns demonstrates the capabilities of IASI to capture the seasonality in HCN and  $\text{C}_2\text{H}_2$  in most cases.

Global seasonal distributions, as well as regional time series of the total columns, have been shown for the two species. IASI is able to capture persisting, seasonal and exceptional features for both species, and the observed patterns are in a general good agreement with previous spaceborne studies (ACE-FTS and MIPAS).

The comparison between these observations and MOZART-4 simulations leads to the following conclusions: (i) the model is able to capture most of the hot spots and seasonal cycles, but not the exceptional events, (ii) the model seems to overestimate (underestimate) anthropogenic (biomass burning) emissions for both species, (iii) the model dynamical scheme and/or the modeled species lifetime could be improved to simulate the impact of the long range transport for these species.

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Global measurements of HCN and  $\text{C}_2\text{H}_2$  from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





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## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Clerbaux, C., Boynard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D., Pommier, M., Razavi, A., Turquety, S., Wespes, C., and Coheur, P.-F.: Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder, *Atmos. Chem. Phys.*, 9, 6041–6054, doi:10.5194/acp-9-6041-2009, 2009.

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**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**V. Dufлот et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**

V. Duflot et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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**Global  
measurements of  
HCN and C<sub>2</sub>H<sub>2</sub> from  
IASI**

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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- 20

## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Table 1.** Global C<sub>2</sub>H<sub>2</sub> and HCN emission sources (Tg(species) year<sup>-1</sup>) during the period 2008–2010 from the dataset used in MOZART-4.

Sources/Year	C <sub>2</sub> H <sub>2</sub>			HCN		
	2008	2009	2010	2008	2009	2010
Anthropogenic	3.37	3.37	3.37	1.67	1.67	1.67
Biomass Burning	0.64	0.71	0.83	1.38	1.33	1.58
Total	4.01	4.07	4.20	3.05	3.00	3.25



## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

**Table 2.** Correlation coefficients ( $R$ ) and biases (Bias) between IASI observations and MOZART-4 simulations for each of the zones defined in Fig. 6.

Zones	C <sub>2</sub> H <sub>2</sub>		HCN	
	$R$	Bias (%)	$R$	Bias (%)
NAM	0.93	47	–	–
NCA	–	–	0.44	–1
SAM	0.54	–50	0.76	–14
EUR	0.88	115	–	–
NAF	0.66	–35	0.07	–2
SAF	0.69	–67	0.86	–28
BCA	0.82	105	–	–
SEA	–0.31	–23	0.57	–4
EQA	0.45	–51	0.09	–27
AUS	0.65	–61	0.77	–26
Global	0.72	–1	0.69	–16

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

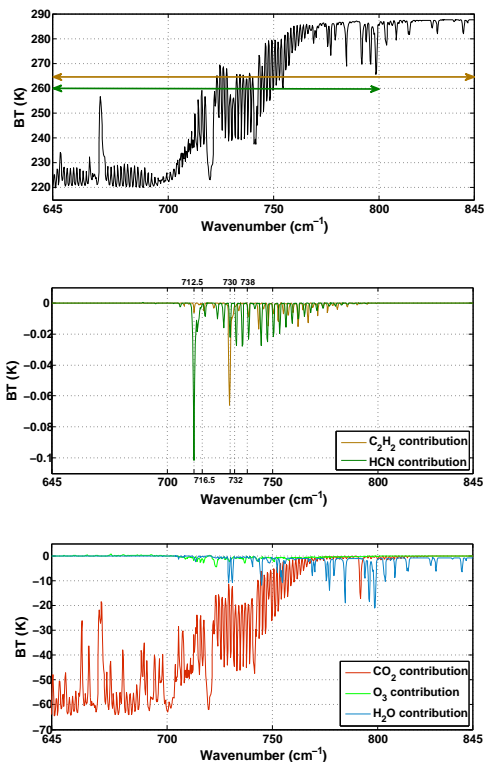
Close

Full Screen / Esc

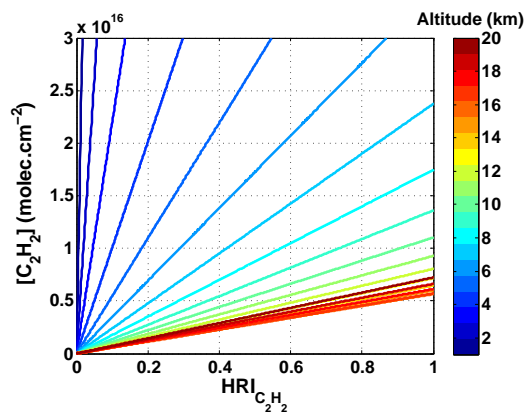
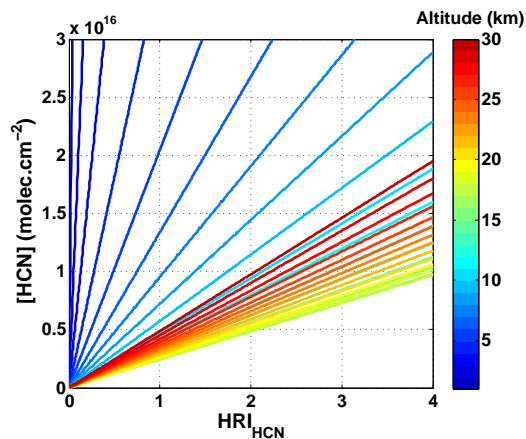
Printer-friendly Version

Interactive Discussion

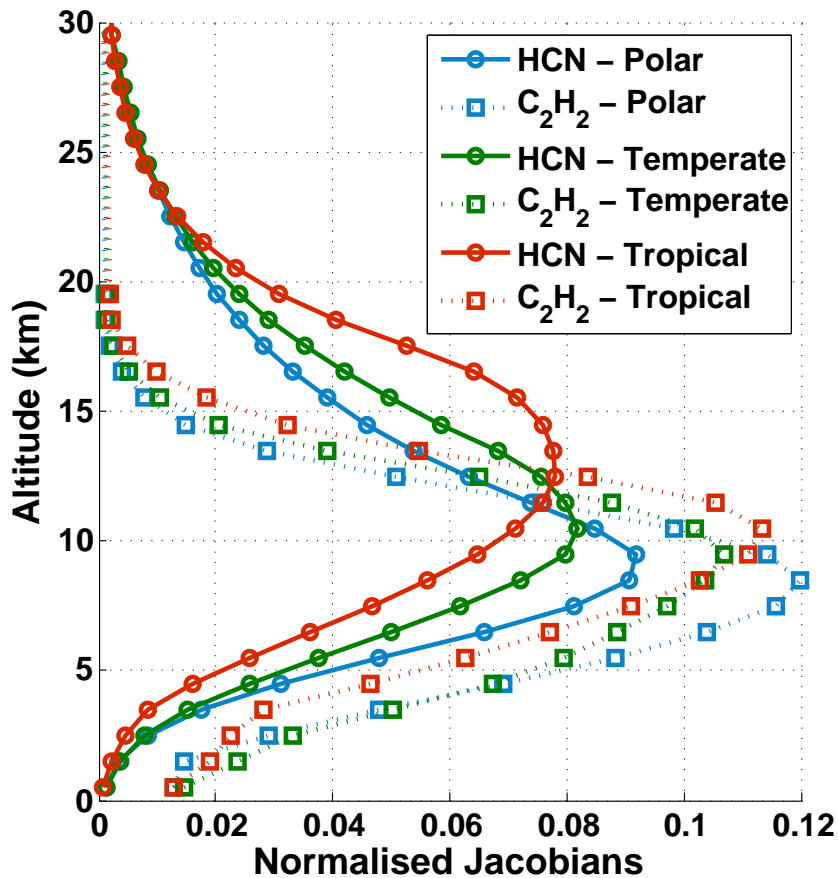




**Figure 1.** (Top) Simulated spectra in the region of the HCN  $\nu_2$  band and  $C_2H_2$   $\nu_5$  band. The green (brown) double sided arrow gives the spectral range used to compute the  $S_e$  matrices for HCN ( $C_2H_2$ ). (Middle) Contributions of climatological background levels of HCN and  $C_2H_2$ . (Bottom) Contribution of  $CO_2$  (red line),  $O_3$  (green line) and  $H_2O$  (blue line) to a simulated spectrum for background concentrations. Calculations have been made for the US Standard Atmosphere (US Government Printing Office, 1976) with  $CO_2$  concentrations scaled to 390 ppmv.



**Figure 2.** Variations of the HRI with HCN (top) and  $C_2H_2$  (bottom) column ( $\text{molec cm}^{-2}$ ) integrated over the 1 km-thick polluted layer in a standard modeled subtropical atmosphere from forward model simulations. The colorscale gives the altitude of the polluted layer.



**Figure 3.** Normalised Jacobians of the forward model implemented in *Atmosphit* for HCN (solid lines) and  $C_2H_2$  (dotted lines) and for the standard modeled polar (blue lines), temperate (green lines) and subtropical (red lines) atmospheres. These are averaged Jacobians over the spectral ranges  $645\text{--}800\text{ cm}^{-1}$  for HCN and  $645\text{--}845\text{ cm}^{-1}$  for  $C_2H_2$ .

Global measurements of HCN and  $C_2H_2$  from IASI

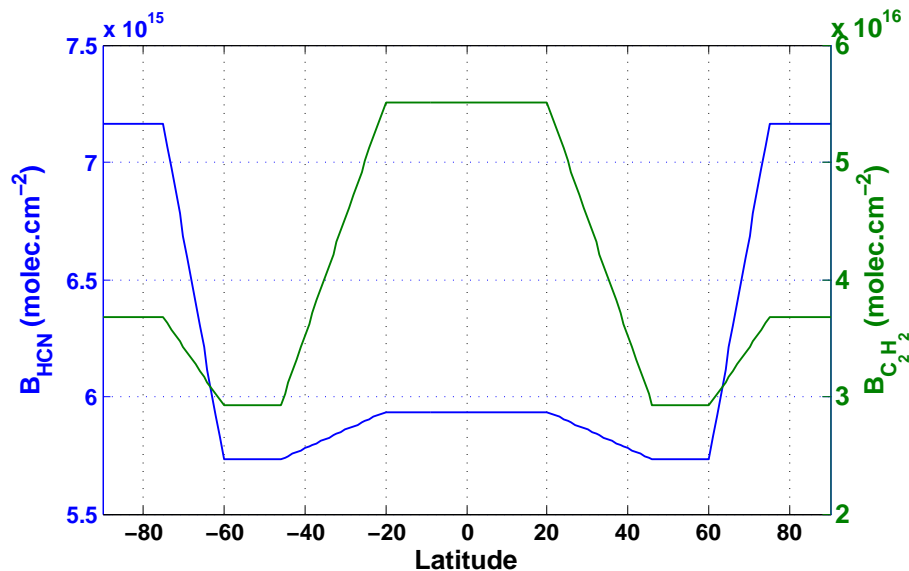
V. Dufлот et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

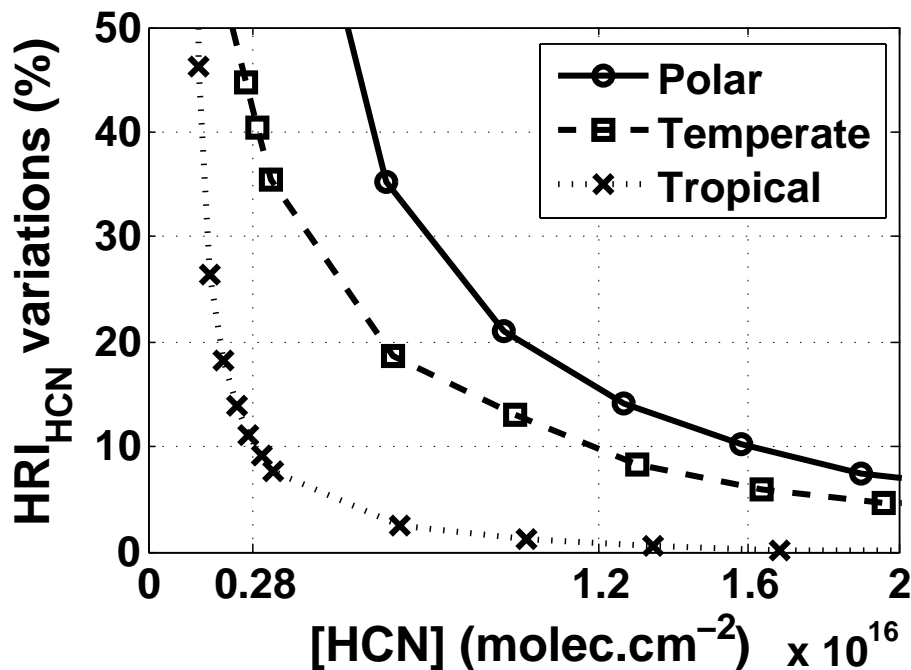


**Figure 4.** Values of  $B_{\text{HCN}}$  (blue) and  $B_{\text{C}_2\text{H}_2}$  (green) as a function of the latitude.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.



**Figure 5.** Variation of  $\text{HRI}_{\text{HCN}}$  caused by a TC equal to  $\pm 10$  K for the polar (solid line and circles), temperate (dashed line and squares) and tropical (dotted line and crosses) atmospheres.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

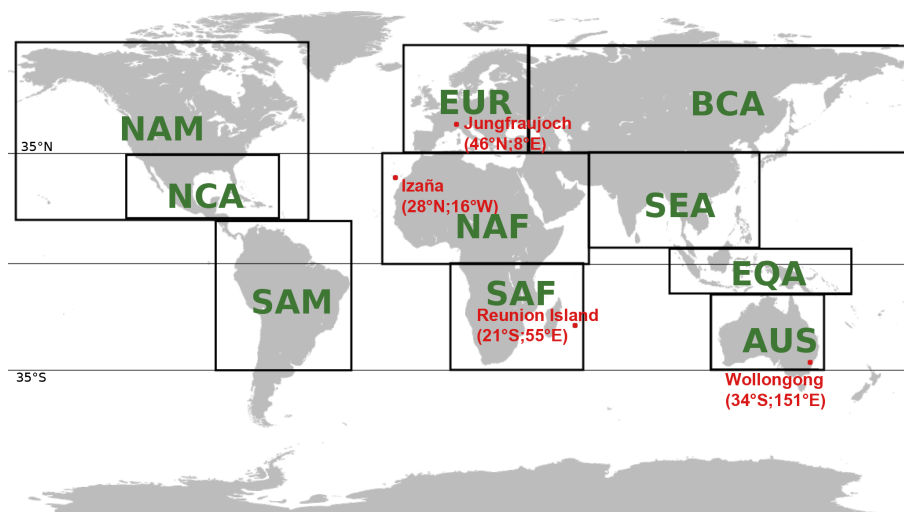
Printer-friendly Version

Interactive Discussion



## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.



**Figure 6.** Locations of the 4 ground-based FTIR measurements sites (Jungfrauoch, Izaña, Reunion Island and Wollongong) and map of the 10 regions used in this study: NAM: Northern America, NCA: North Central America, SAM: South America, EUR: Europe, NAF: Northern Africa, SAF: Southern Africa, BCA: Boreal Central Asia, SEA: South East Asia, EQA: Equatorial Asia, AUS: Australia.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

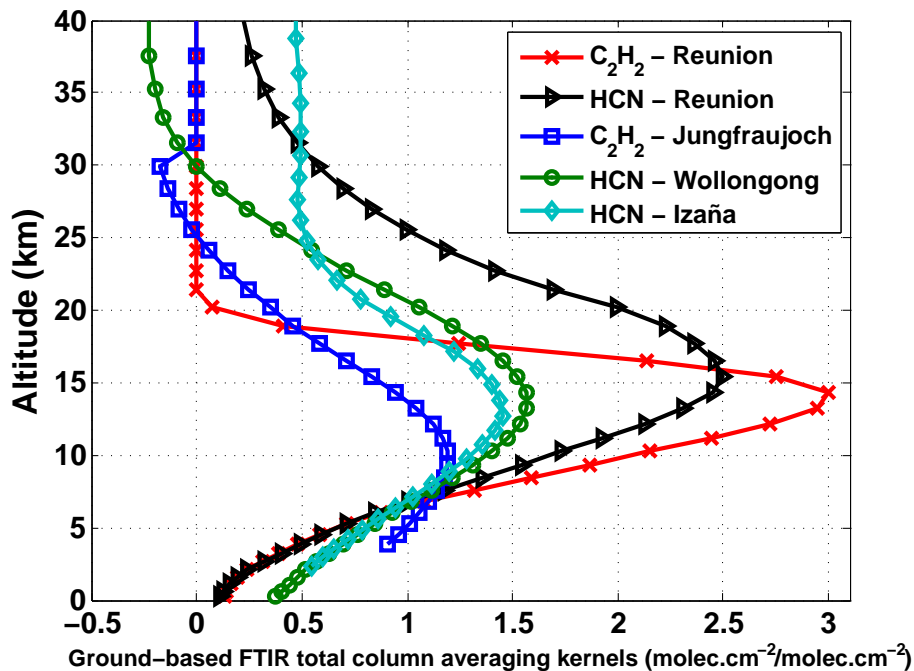
Printer-friendly Version

Interactive Discussion



Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.



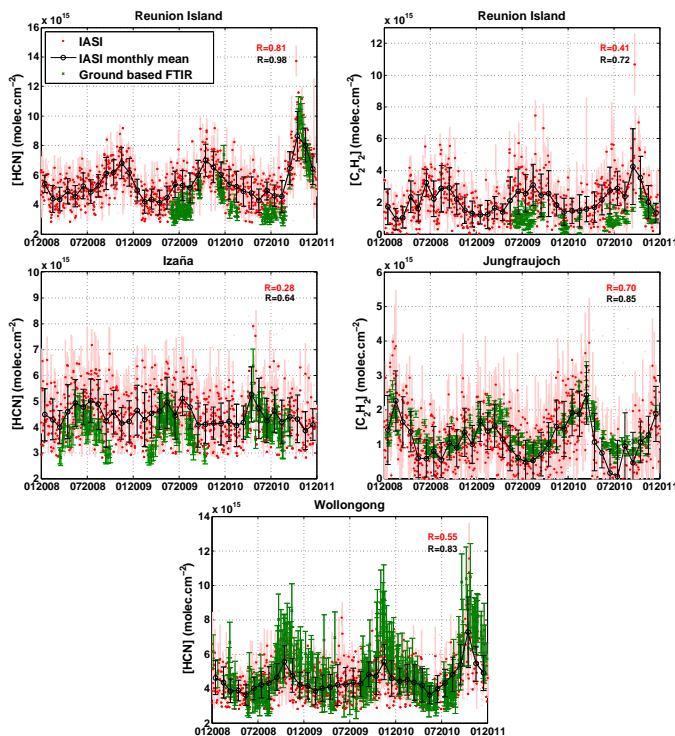
**Figure 7.** Total column averaging kernels of ground-based FTIR in  $\text{molec cm}^{-2}/\text{molec cm}^{-2}$  for C<sub>2</sub>H<sub>2</sub> at Reunion Island (red stars and line), HCN at Reunion Island (black circles and line), HCN at Wollongong (green dots and line), HCN at Izaña (light blue diamonds and line) and C<sub>2</sub>H<sub>2</sub> at Jungfrauoch (blue squares and line).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

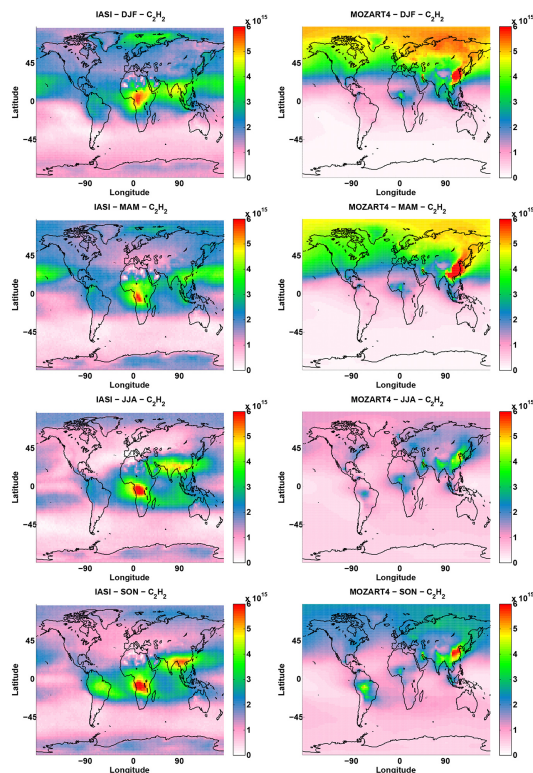
V. Dufлот et al.



**Figure 8.** Time series of HCN (left panel) and C<sub>2</sub>H<sub>2</sub> (right panel) measurements for Reunion Island (HCN and C<sub>2</sub>H<sub>2</sub>), Wollongong (HCN only), Izaña (HCN only), and Jungfraujoeh (C<sub>2</sub>H<sub>2</sub> only). IASI measurements are shown as daily and 1° × 1° means (red dots) with associated SDs (light red lines), and as monthly and 1° × 1° means (black circles and line) with associated SD (vertical black lines). Ground-based FTIR measurements are shown as daily means with associated total error by green crosses and lines. Correlation coefficients are given on each plot for daily means in red and for monthly means in black.

## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.



**Figure 9.** Seasonal distribution of the C<sub>2</sub>H<sub>2</sub> total column (in molec cm<sup>-2</sup>) as measured by IASI (left panel) and simulated by MOZART-4 (right panel) averaged over the years 2008 to 2010. The IASI global distributions are given with the same horizontal resolution as MOZART-4 (1.875° latitude × 2.5° longitude). DJF = December–January–February, MAM = March–April–May, JJA = June–July–August, SON = September–October–November.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

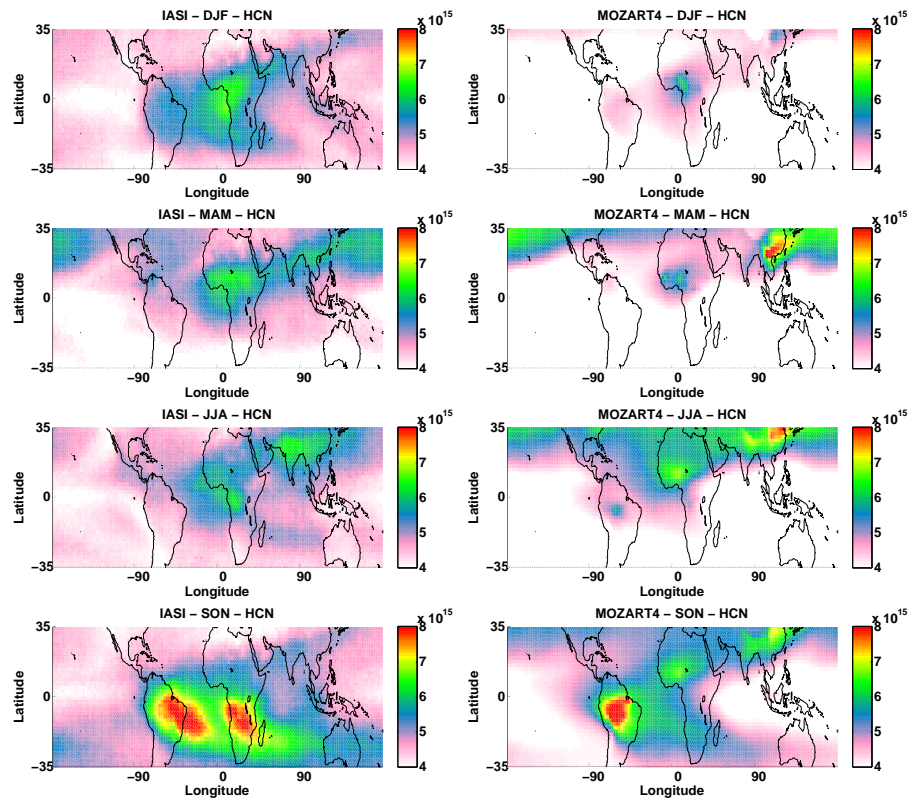
[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Figure 10. Same as Fig. 9 for HCN.

## Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

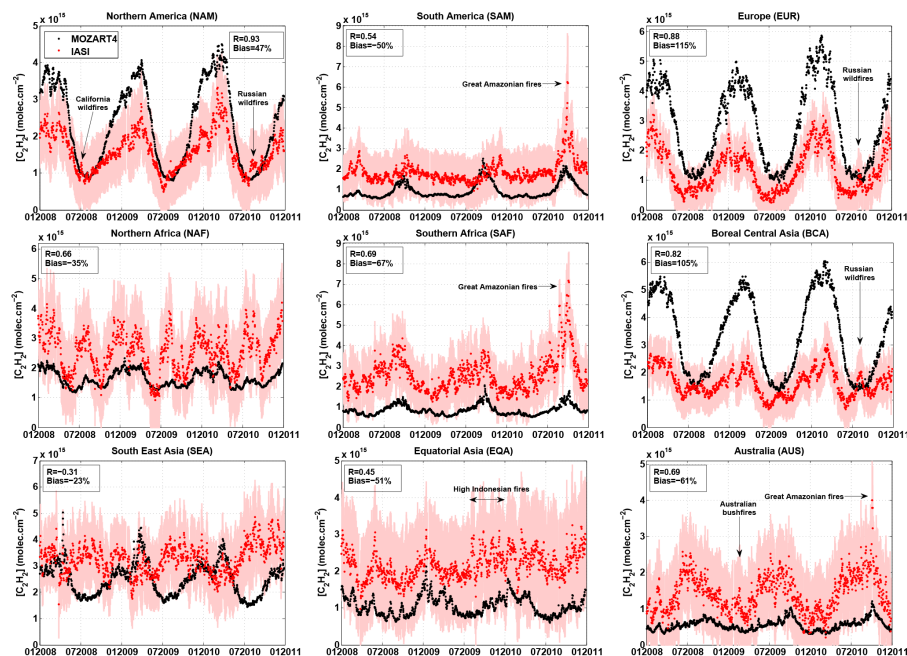
Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Figure 11.** Evolution with time of the mean C<sub>2</sub>H<sub>2</sub> total column (in molec cm<sup>-2</sup>) over the zones defined in Fig. 6 as measured by IASI (red dots) with associated SD (light red lines), and as simulated by MOZART-4 (black dots). Correlation coefficients ( $R$ ) and biases (Bias) between IASI and MOZART-4 are given on each plot for daily means.

Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Dufлот et al.

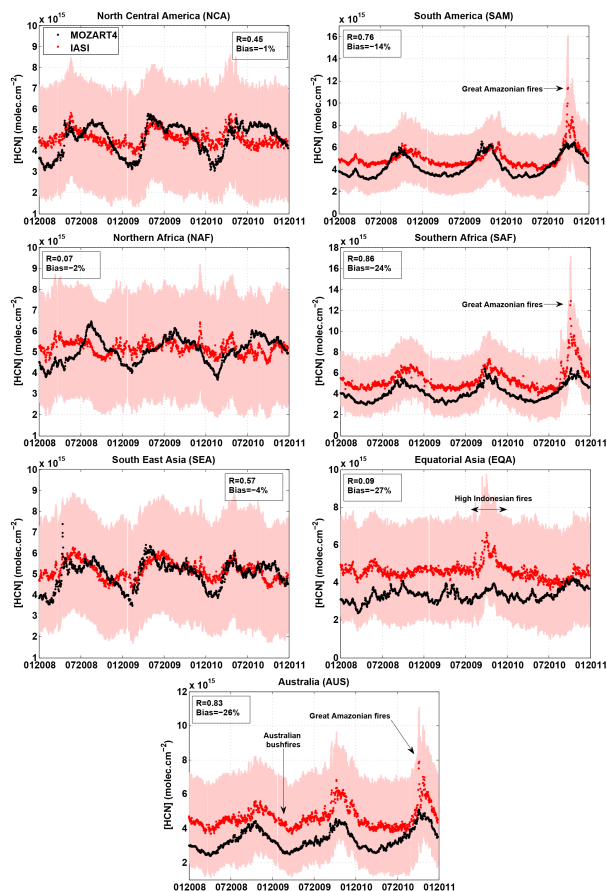


Figure 12. Same as Fig. 11 for HCN.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global measurements of HCN and C<sub>2</sub>H<sub>2</sub> from IASI

V. Duflot et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

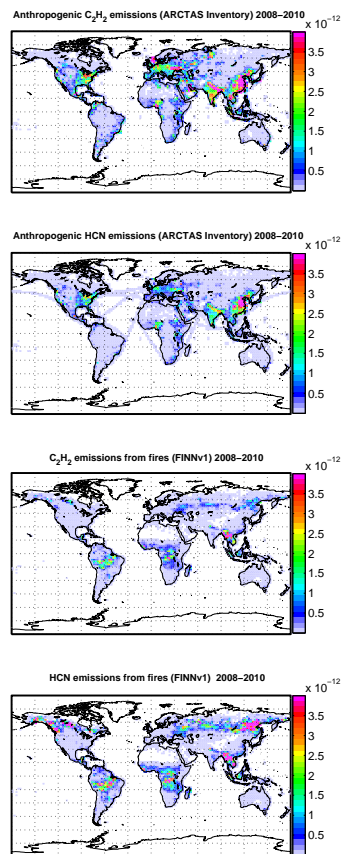
Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Figure 13.** C<sub>2</sub>H<sub>2</sub> and HCN surface emission fluxes (kg m<sup>-2</sup> s<sup>-1</sup>) averaged over the period 2008–2010 from the anthropogenic and fire emissions inventories used in MOZART-4.