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Thermal infrared nadir observations of 24 atmospheric gases

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[1] Thermal infrared nadir sounders are ideal for observing total columns or vertical profiles of atmospheric gases such as water, carbon dioxide and ozone. High resolution sounders with a spectral resolution below 5 cm^{-1} can distinguish fine spectral features of trace gases. Forty years after the launch of the first hyperspectral sounder IRIS, we have now several state of the art instruments in orbit, with improved instrumental specifications. In this letter we give an overview of the trace gases which have been observed by infrared nadir sounders, focusing on new observations of the Infrared Atmospheric Sounding Interferometer (IASI). We present typical observations of 14 rare reactive trace gas species. Several species are reported here for the first time in nadir view, including nitrous acid, furan, acetylene, propylene, acetic acid, formaldehyde and hydrogen cyanide, observations which were made in a pyrocumulus cloud from the Australian bush fires of February 2009. Being able to observe this large number of reactive trace gases will likely improve our knowledge of source emissions and their impact on the environment and climate. **Citation:** Clarisse, L., Y. R'Honi, P.-F. Coheur, D. Hurtmans, and C. Clerbaux (2011), Thermal infrared nadir observations of 24 atmospheric gases, *Geophys. Res. Lett.*, 38, L10802, doi:10.1029/2011GL047271.

1. Introduction

[2] The first high resolution terrestrial infrared spectra were measured from space by the Infrared Interferometer Spectrometer (IRIS, 1969–1971) on-board the meteorological platform Nimbus 3 and 4, revealing carbon dioxide, water vapor, methane, nitrous oxide and ozone absorption bands [Hanel *et al.*, 1972]. Although the potential of nadir looking hyperspectral infrared sounders was realized early on, the technology at the time could not provide the spatial resolution and revisit time required for the operational retrieval of weather variables. In the following decades, preference was given to broadband infrared sounders for measuring clouds, temperature and humidity. In the 1990s, the climate change debate spurred the development of the Interferometric Monitor for Greenhouse Gases (IMG, 1996–1997). It was designed to measure a host of trace gases such as CO_2 , H_2O , O_3 , CO and CH_4 [Shimoda and Ogawa, 2000; Clerbaux *et al.*, 2003]. A comparison between IRIS and IMG spectra revealed an increase in greenhouse gases absorptions, including for CFC-11 and CFC-12 [Harries *et al.*, 2001].

[3] IMG died prematurely, but fortunately since 2002 new nadir-looking high resolution spectrometers have been launched, missions which are motivated by increasing demands on numerical weather forecasting and on monitoring atmospheric composition. These are Atmospheric Infrared Sounder (AIRS, 2002- [Chahine *et al.*, 2006]), Tropospheric Emission Spectrometer (TES, 2004- [Beer, 2006]), Infrared Atmospheric Sounding Interferometer (IASI, 2006-) and Greenhouse Gases Observing Satellite (GOSAT, 2009- [Kuze *et al.*, 2009]). In this letter we focus on IASI, which is arguably the most versatile of these. IASI is cross track scanner with 4 pixels and a swath of 2400 km on board the sun-synchronous polar-orbiting Metop-A platform. It makes about 1280000 measurements per day of the Earth's outgoing infrared radiation, adding up to twice daily global coverage. Its footprint ranges from circular with a 12 km diameter at nadir to roughly elliptical with axes of 39 km (across track) and 20 km (along track) at the swath maximum. The spectrometer has an apodized spectral resolution of 0.5 cm^{-1} , spectral coverage extending from $645\text{ to }2760\text{ cm}^{-1}$ ($3.6\text{ to }15.5\text{ }\mu\text{m}$) and low radiometric noise ($\text{Ne}\Delta\text{T} \sim 0.2\text{ K}$ below 2300 cm^{-1} and $>0.3\text{ K}$ above) [Clerbaux *et al.*, 2009].

[4] There are ten species which can be observed in each IASI spectrum, namely CO_2 , N_2O , CFC-11, CFC-12, OCS, H_2O , CH_4 , O_3 , CO and HNO_3 [see Clerbaux *et al.*, 2009, and references therein]. All these are omnipresent and have clear spectral signatures. Measuring their background abundances and local variations is important for monitoring and studying tropospheric and stratospheric chemistry and climate.

[5] In recent years, a number of (mostly short-lived) trace gases have been discovered in IASI spectra. These are sulfur dioxide (SO_2) [Clarisse *et al.*, 2008], ammonia (NH_3), methanol (CH_3OH), formic acid (HCOOH), ethene (C_2H_4) and PAN ($\text{C}_2\text{H}_3\text{O}_5\text{N}$) [Coheur *et al.*, 2009]. Enhanced loadings above or downwind from a strong emission source are usually required for these to be observable. This can be an extreme point source such as a volcano or a large wildfire, with possible injection heights up in stratosphere. Other sources typically have a lower emission height, as the source is less violent and covers a larger area (e.g., biogenic emissions from plants, anthropogenic emissions from intensive agriculture or megacities). Given sufficient loadings and adequate atmospheric conditions these emissions can be observed in the boundary layer, or via uplift in the lower troposphere. For some species (NH_3 , CH_3OH and HCOOH) global or quasi-global distributions could be obtained [Clarisse *et al.*, 2009; Razavi *et al.*, 2011].

[6] IASI is now almost four years in orbit, and in this letter we give an overview of the 14 rare reactive trace gases which have been observed so far through the presentation of example spectra (we do not discuss the 10 species observable in each spectrum here). We report several species

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which have never been observed before with a nadir sounder. These were identified in a pyrocumulus cloud after the Australian bush fires of February 2009 [Trepte *et al.*, 2009]. For the species which were detected before, we present extreme examples. Here we deal with the detection of trace gases, a detailed quantitative study and analysis of, e.g., in-plume chemistry, is outside the scope of this letter. Also note that some of the species have been detected before by limb sounders [Coheur *et al.*, 2007].

2. Global Observations

[7] The main result of the paper is Figure 1, which presents the observed spectra of the 14 species. These are illustrated in two different ways. When the spectral signature can be distinguished clearly from other interfering species, the raw spectrum is displayed (in black), with the molecular signature superimposed (in red). As we deal with short lived trace gases, this was only possible in a few cases (C_2H_4 , NH_3 , SO_2 and CH_3OH). For the other species we used the optimal estimation technique to make a synthetic reconstruction of the spectrum. The residual, i.e., the difference between observed and fitted spectrum represents ideally the instrumental noise. As done by Coheur *et al.* [2009] we excluded specific species in the reconstructed spectrum, and are therefore able to show the presence of their signature (red) in the residual (blue). The forward and inverse calculations were done with Atmosphit [Coheur *et al.*, 2005] using the HITRAN spectroscopic database [Rothman *et al.*, 2009]. For species such as HONO which are absent in HITRAN, we used the PNNL infrared cross sections database [Sharpe *et al.*, 2004]. Recently global distributions have been obtained for NH_3 , CH_3OH , $HCOOH$; these species will be discussed in this section.

[8] The ammonia spectrum shown in Figure 1 was observed over Buenos Aires Province, Argentina on 18 April 2008. That day Buenos Aires made the world news as it was blanketed with thick smoke from inland burning of grass and shrubs. The spectrum is quite unusual as ammonia is present here in emission, with signatures visible above the baseline, indicating that the air temperature in the fire plume was raised relative to the surface temperature. It is illustrative for the type of conditions where ammonia is observable. As a short lived species it is mainly confined to the boundary layer, and to observe it, a medium to large temperature gradient is needed between the surface and the first atmospheric layer. This can be a negative gradient (as shown here), but more commonly, a positive gradient. For the San Joaquin Valley in California for instance, which is known for its intensive agriculture, a recent study demonstrated that IASI was able to measure NH_3 for the eight hottest months [Clarisse *et al.*, 2010].

[9] The volatile organic compounds methanol and formic acid are among the most important organic species in our atmosphere. Their estimated emissions are respectively of the order of 200 and 10 $Tg\ yr^{-1}$ and are primarily biogenic in origin. The strongest signatures were found in fire plumes though, when local concentrations can be very high. For methanol a spectrum is shown over the Australian fire plume in 2009 and for formic acid over a fire in Botswana in October 2008. While the signatures are obvious here, routine detection is hampered by strongly interfering ozone (for methanol) and water lines (for formic acid). With a

lifetime of around a week, global observations are possible for both species, revealing emission sources and transport patterns [Razavi *et al.*, 2011]. From these results, methanol emissions have been obtained, leading to an improved model agreement with independent measurements [Stavrakou *et al.*, 2011].

3. Local Observations

[10] This section discusses the species which are observed sporadically, such as SO_2 and H_2S after a volcanic eruption. While UV instruments have the longest heritage in SO_2 sounding [Torres *et al.*, 2002], it has been known for some-time that infrared instruments are also capable of measuring atmospheric SO_2 [Prata *et al.*, 2003]. The SO_2 spectrum shown in Figure 1 was observed after the volcanic eruption of Kasatochi in August 2008 [Karagulian *et al.*, 2010]. Three distinct absorption bands can be distinguished. The band associated with the ν_3 vibrational mode is the strongest, however, interfering water vapour makes this band only sensitive to SO_2 above the mid-troposphere. Most observations made in this band are volcanic plumes, although the occasional uplift of anthropogenic SO_2 is observed too. The ν_1 and $\nu_1 + \nu_3$ bands are much weaker, but have a larger sensitivity to lower tropospheric SO_2 and can also be useful in case the ν_3 band saturates.

[11] First observations of H_2S were recently made in the plume of the Kasatochi eruption and an example spectrum is shown in Figure 1 (the event is discussed in detail by Clarisse *et al.* [2011], which presents also more detailed spectroscopic evidence). This identification of hydrogen sulfide was possible because of IASI's very low noise in this spectral region (~ 0.04 K) and height of the plume. The detection limit for an upper tropospheric plume has been estimated at 25 DU. Such an initial observation is important, as there are large uncertainties in the contribution of H_2S to the global sulfur budget. It also has importance in terms of remote detection of eruption style (magmatic/phreatomagmatic).

[12] The remaining species are typical products from biomass burning for which the examples were taken from the 2009 Australian bushfires. Three species, namely hydrogen cyanide (HCN), furan (C_4H_4O) and acetylene (C_2H_2) were observed in the very strong ν_2 band of CO_2 between 500 and $800\ cm^{-1}$. It is only thanks to their particularly strong signature that unambiguous identification was possible. For HCN and C_4H_4O this is a Q-branch which exceeds 5 K at the top of the atmosphere. The acetylene signature shows up strongly in the ν_5 -Q branch, but some of the R lines are clearly visible as well. At shorter wavelengths propylene (C_3H_6) is observed in its ν_{19} band around $912\ cm^{-1}$, and while the synthetic fit is far from perfect, the signature clearly emerges from the residual.

[13] Formaldehyde (HCHO) and especially nitrous acid (HONO) are important precursors of the hydroxyl radical (OH), which to a large extent determines the oxidation capacity and thus rate of removal of most pollutants [Elshorbany *et al.*, 2009]. Both were observed in the Australian fire plume. While formaldehyde is readily observed from space in the ultraviolet [De Smedt *et al.*, 2008], the observation of nitrous acid presented here is the first from space and in view of its strong signature, future observations can be expected. The residual of formaldehyde only barely emerges from the instrumental noise, but shows up

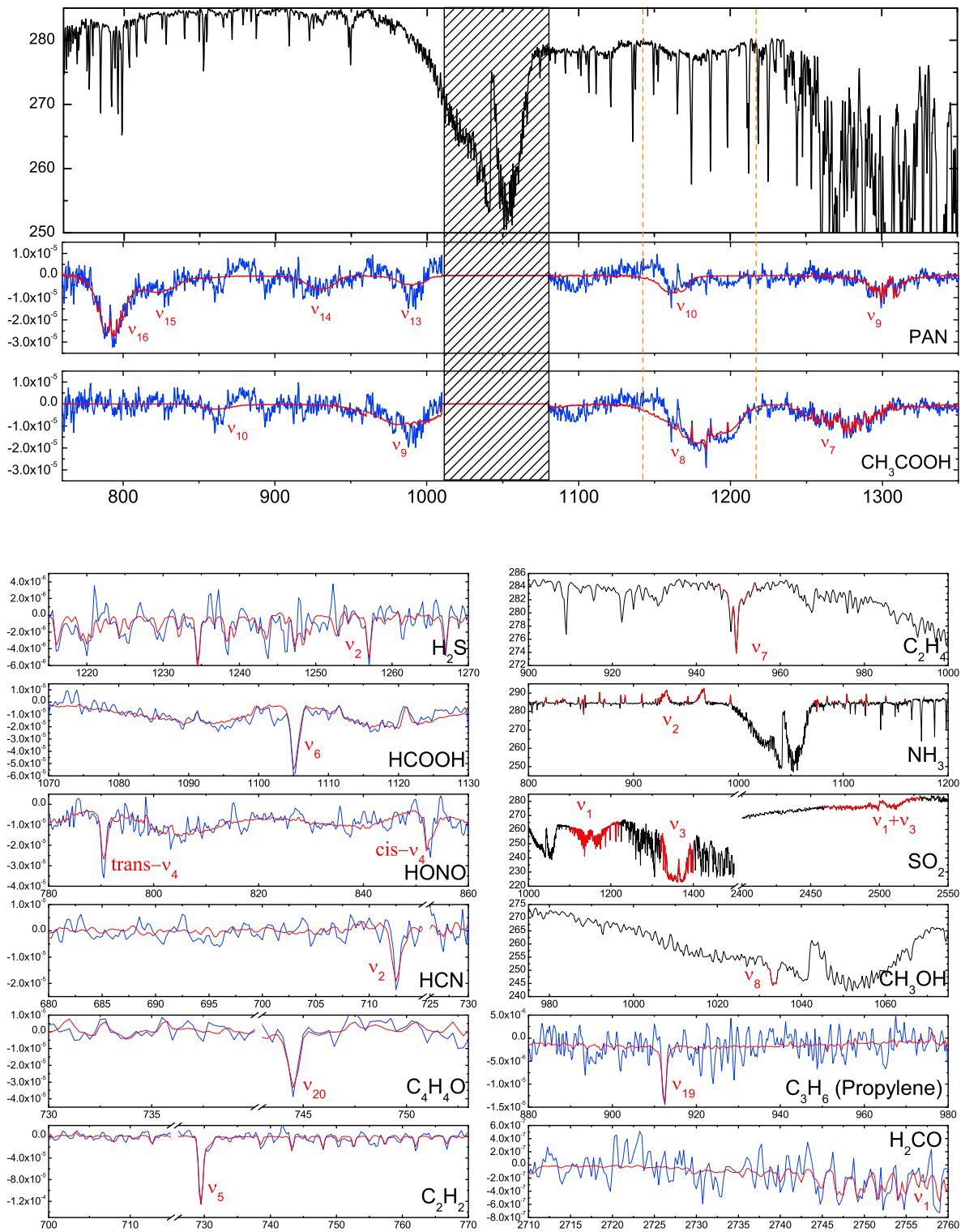


Figure 1. Observed IASI spectra of volcanic and pyrogenic events revealing 14 rare trace gas species. The spectra in black are in brightness temperature in units Kelvin, while the spectra in blue are in radiance in units $\text{Wm}^{-2}\text{m Sr}^{-1}$, the horizontal axes are in cm^{-1} units.

unambiguously when making the average of several spectra in the smoke plume.

[14] Ethylene (C_2H_4), acetic acid (CH_3COOH) and PAN were observed by IASI before [Coheur *et al.*, 2009]. The

ethylene signature is exceptionally strong, and easily distinguishable in the spectra of many fire plumes. The different absorption bands of CH_3COOH and PAN observations are shown in the residual in Figure 1. Previously only the PAN

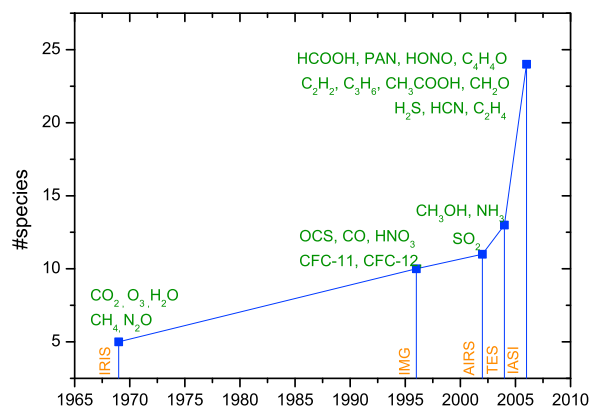


Figure 2. Historical overview of cumulative detected species from infrared nadir sounders: IRIS [Hanel et al., 1972], IMG [Clerbaux et al., 2003], AIRS [Carn et al., 2005], TES [Beer et al., 2008] and IASI [Coheur et al., 2009; this paper]. Because the exact date of first detection is hard to determine, the species are listed by launch date of the respective instruments.

band at 794 cm^{-1} had been observed, while the acetic acid assignment was only tentative. Some of the absorption bands are partially overlapping, such as those in the region between 1140 and 1220 cm^{-1} (marked in orange), and could falsely be attributed to an other molecular absorption if a larger part of the spectrum is not analysed. Figure 1 also shows the observed spectrum, to illustrate its rich structure. Apart from the many different molecular absorptions, an aerosol contribution can also be distinguished as a broadband drop in the baseline (the background surface temperature is around 295 K) and as a large absorption in the region $1050\text{--}1250\text{ cm}^{-1}$.

4. Conclusion

[15] In this letter we have presented spectroscopic evidence of 14 rare trace gases in IASI spectra. This includes seven species which have never been reported before with a nadir sounder. Figure 2 illustrates the progress of nadir IR remote sensing in measuring reactive trace gases, since the first observations of IRIS in 1969. The recent steep increase in detected species is not so much due to an increase in spectral resolution as it is to improved radiometric performances and an improved spatial and temporal coverage (more events are detected, and detected early on). IASI will be in orbit for another 10 years (on Metop-B and Metop-C), and follow-up missions with improved instruments are already in the pipeline. The potential for observing this large number of trace gases is therefore important and is anticipated to lead to many advances in our understanding of source emissions, atmospheric chemistry and impacts on environment and climate.

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