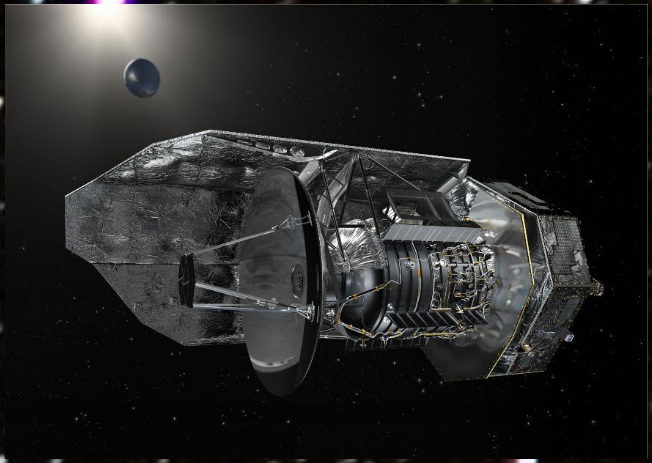
Measurement of isotopic ratios in solar system bodies



Nicolas Biver,
LESIA, CNRS,
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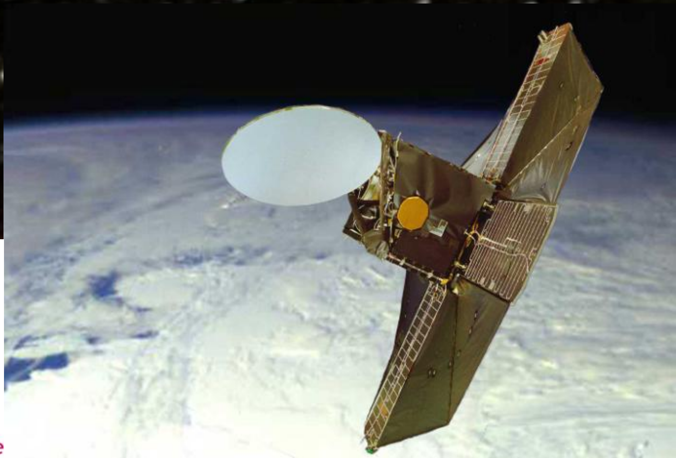

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Laboratoire d'Études Spatiales et d'Instrumentation en Astrophysique

Measuring isotopic ratios in solar system objects

Measurements in cometary, planetary atmospheres :

• <u>In situ measurements</u> (mass spectrometry (MS), *e.g. Giotto, Rosetta, Huygens, Cassini*)

• <u>Laboratry measurements</u>: sample return (*e.g. 81P/Wild 2 - Stardust*) or meteorites

• <u>UV or visible spectroscopy</u> : radicals (CN, C_2, OH, NH_2), atoms (D)

• <u>InfraRed and radio spectroscopy</u> : molecules (HDO, $H_2{}^{18}O$, $HC^{15}N$, $H_2{}^{34}S$, HD, CH_3D,…) radicals ($C^{34}S$): remote (*JCMT,IRAM, ALMA,VLT, KecK, Subaru, Herschel…*) and in-situ (*VIRTIS,MIRO-Rosetta, CIRS-CASSINI*) – narrow comet lines (2km/s), broad planetary lines (several GHz)

⇒***All spectroscopy techniques require large telescopes and high spectral resolution to detect and separate the weak lower abundance isotopologue***

<u>Review papers :</u>

• *Jehin et al. 2009 (EMP, 105, 167)*

• ***Bockelée-Morvan et al. 2015 (Space science Review 197, 47-83)***

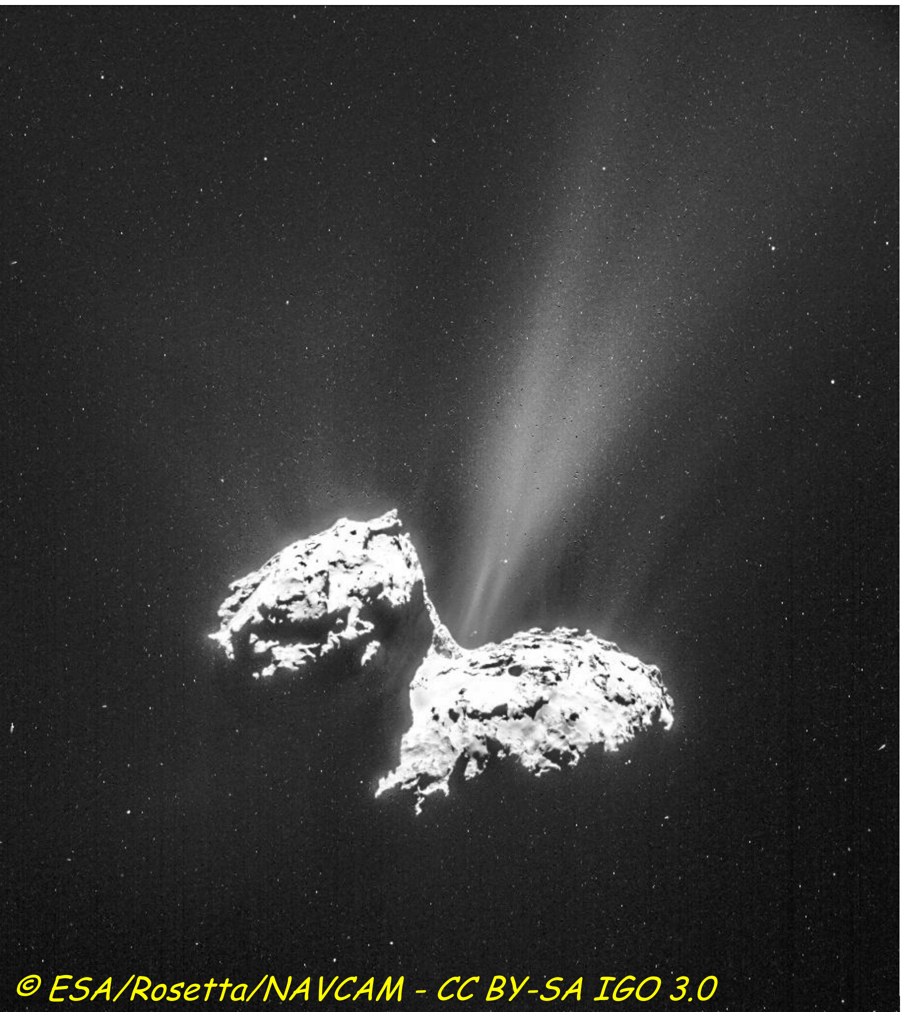
• *Bézard et al. 2014 (in « Titan, interior, Surface, Atmosphere and Space Environment », Eds I. Müller-Wodarg, C.A. Griffith, E. Lellouch and T. Cravens - Cambridge Planetary Science)*

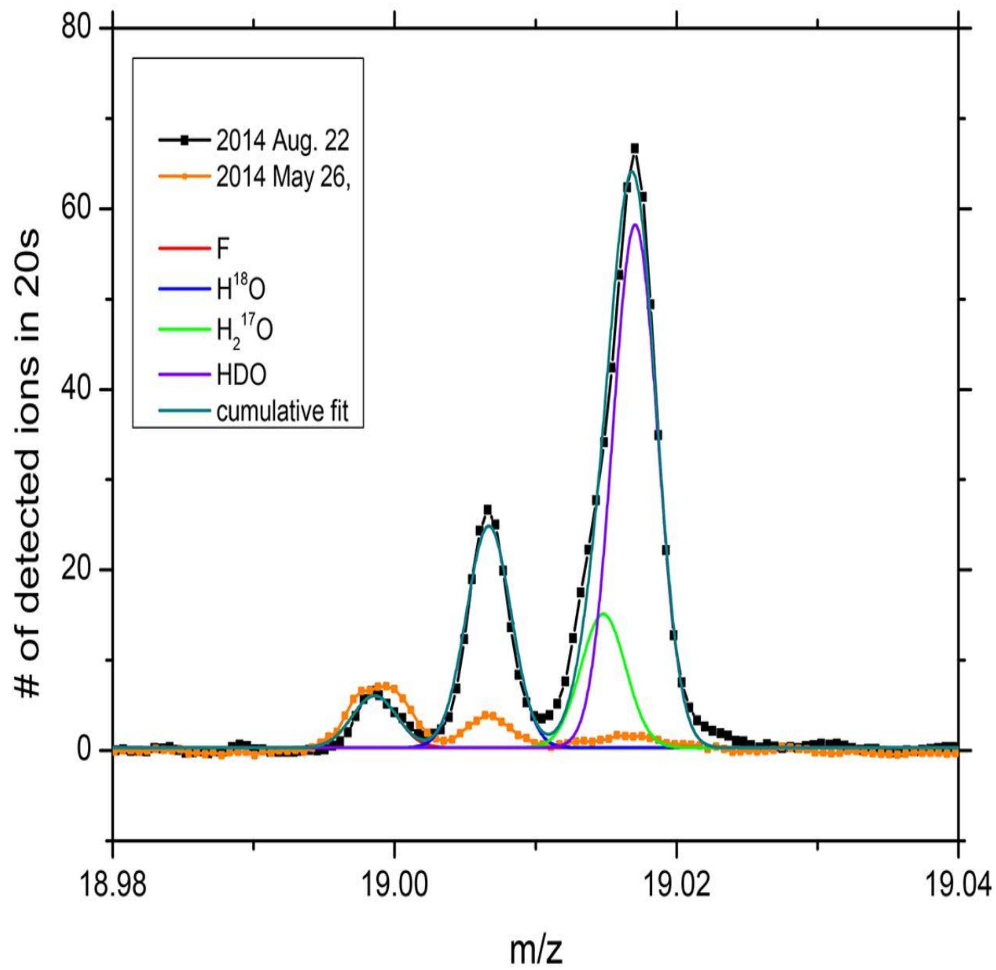
• *Füri and Marty 2015, Nature Geoscience 8, 515-522*

Mass Spectrometry: HDO in comet 67P/C.G.

Rosina/DFMS: high mass resolution spectrometer needed to separate various element of close m/z ratio

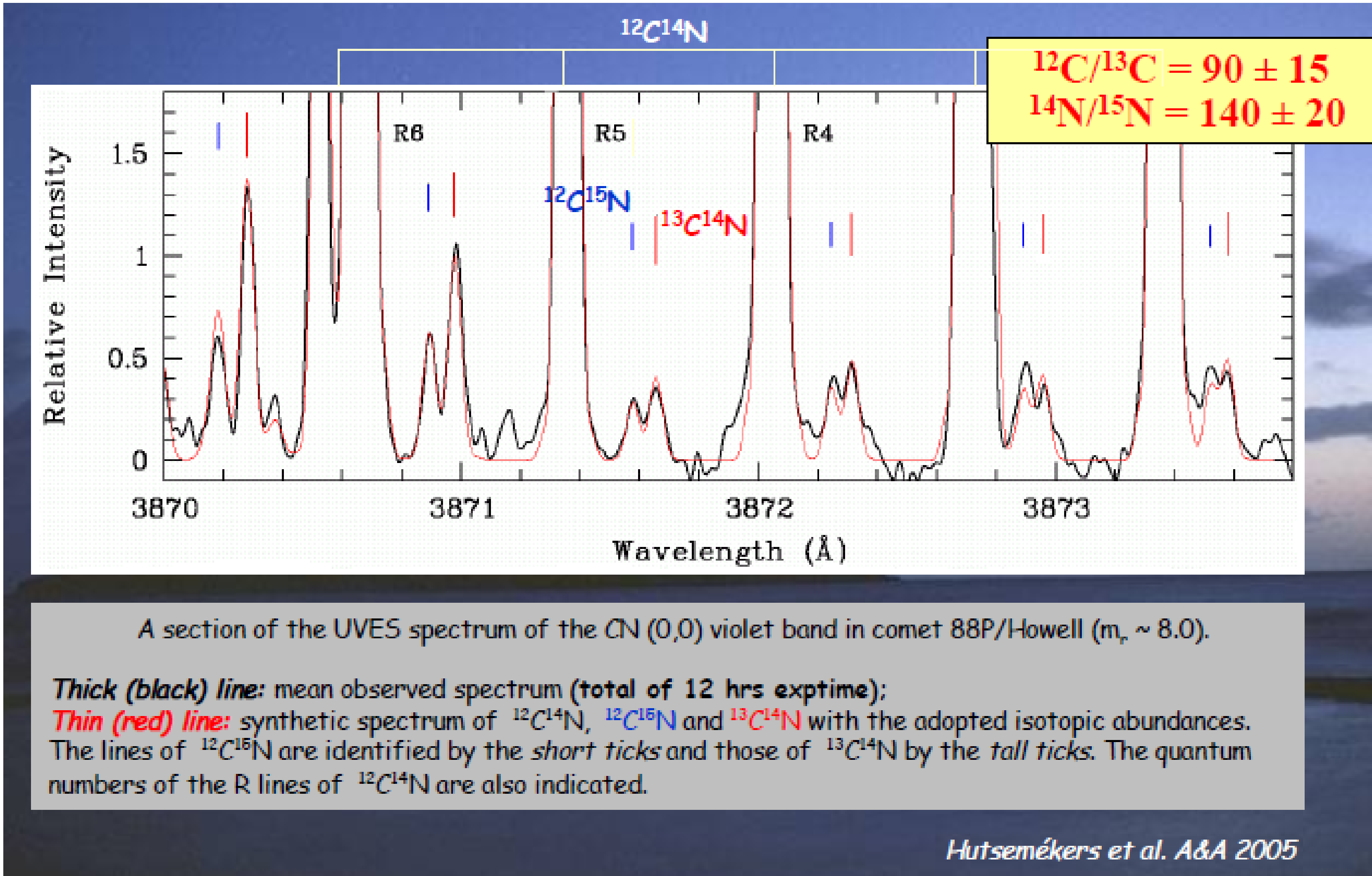
Altwegg et al. 2015, Science 347, 1261952





UV or Visible spectroscopy:

Individual rovibrational transitions within the electronic bands:
high spectral resolution required (>30000)



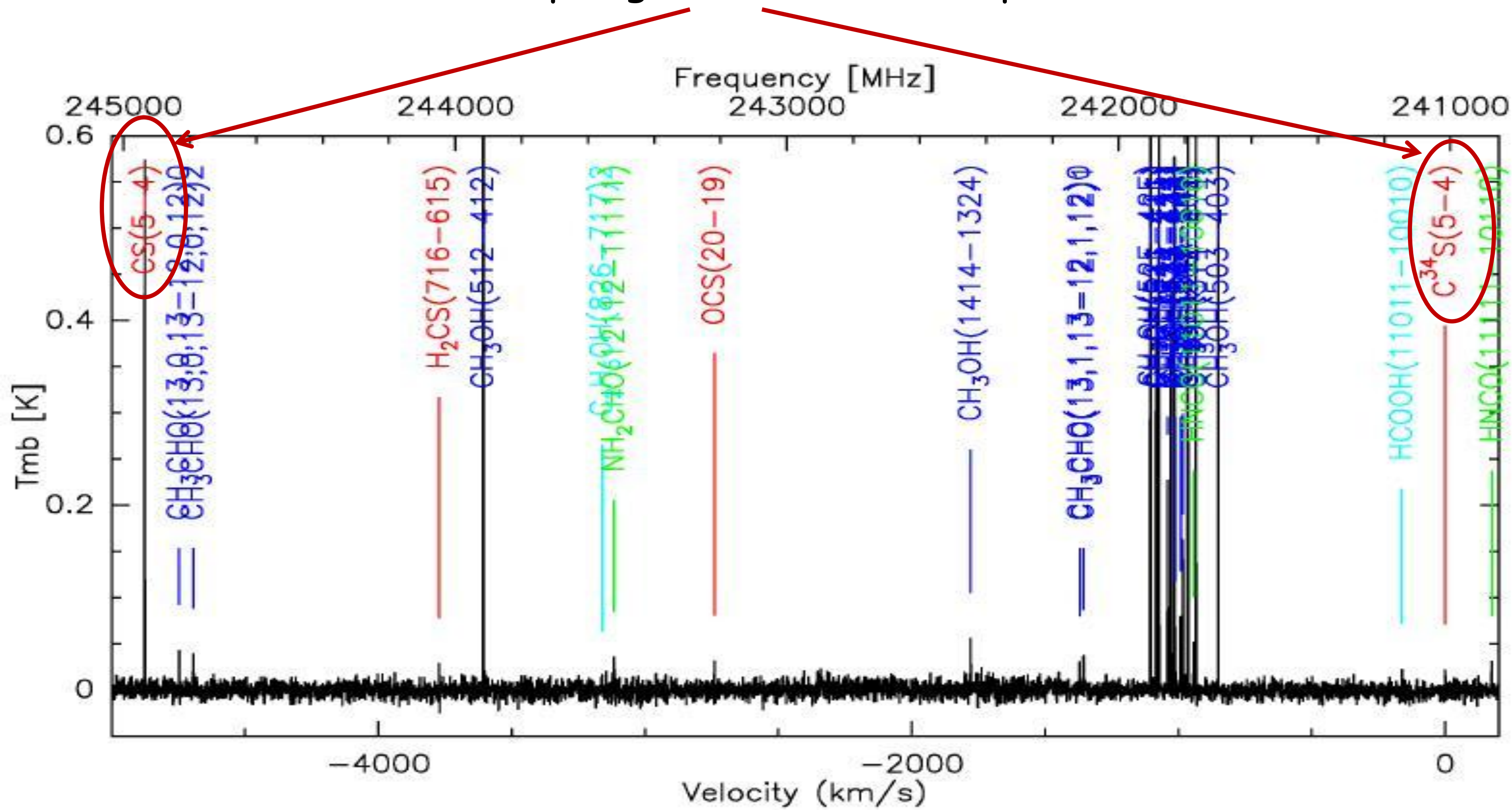
A section of the UVES spectrum of the CN (0,0) violet band in comet 88P/Howell (m_r ~ 8.0).

Thick (black) line: mean observed spectrum (**total of 12 hrs exptime**);
Thin (red) line: synthetic spectrum of $^{12}C^{14}N$, $^{12}C^{15}N$ and $^{13}C^{14}N$ with the adopted isotopic abundances.
The lines of $^{12}C^{15}N$ are identified by the *short ticks* and those of $^{13}C^{14}N$ by the *tall ticks*. The quantum numbers of the R lines of $^{12}C^{14}N$ are also indicated.

Hutsemékers et al. A&A 2005

Radio spectroscopy:

high sensitivity/strong source required and wide frequency coverage:
Isotopologues several GHz apart



C/2014 Q2 (Lovejoy): IRAM 30m 13-25 Jan. 2015

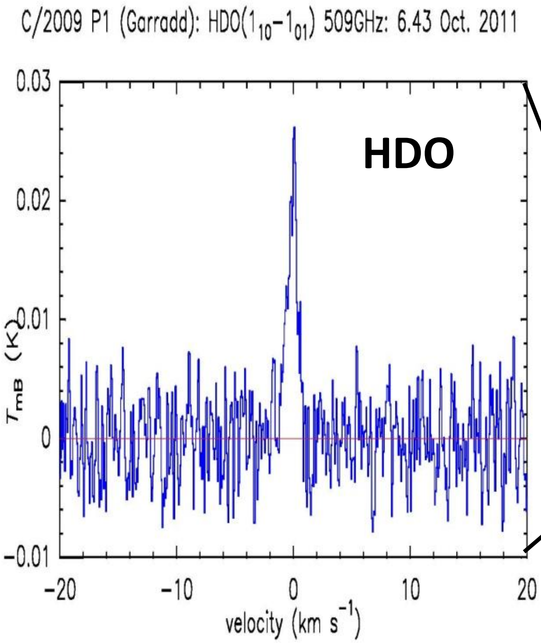
Radio spectroscopy: (Herschel)

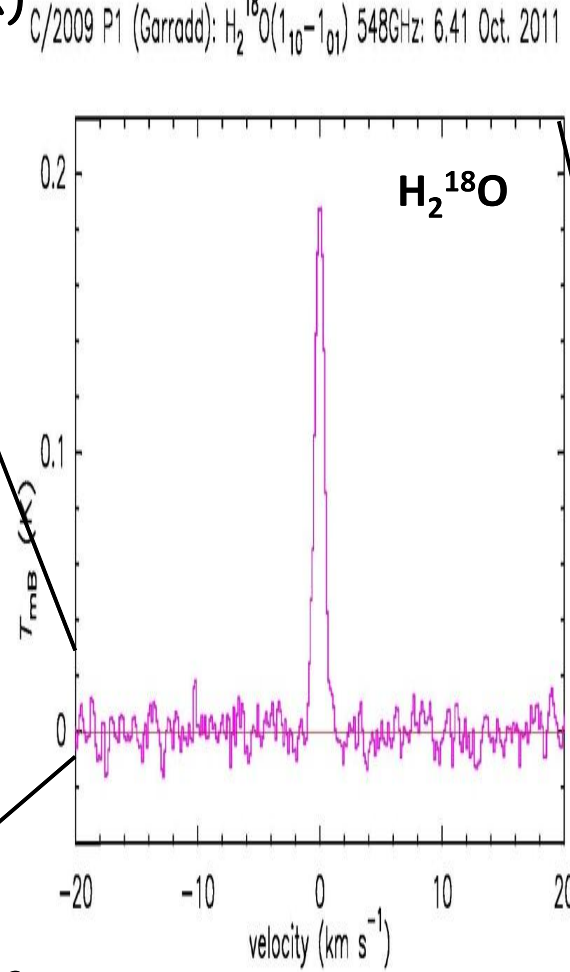
Observations of HDO, $H_2^{18}O$ and $H_2^{16}O$ at 509-557 GHz with HIFI/Herschel:

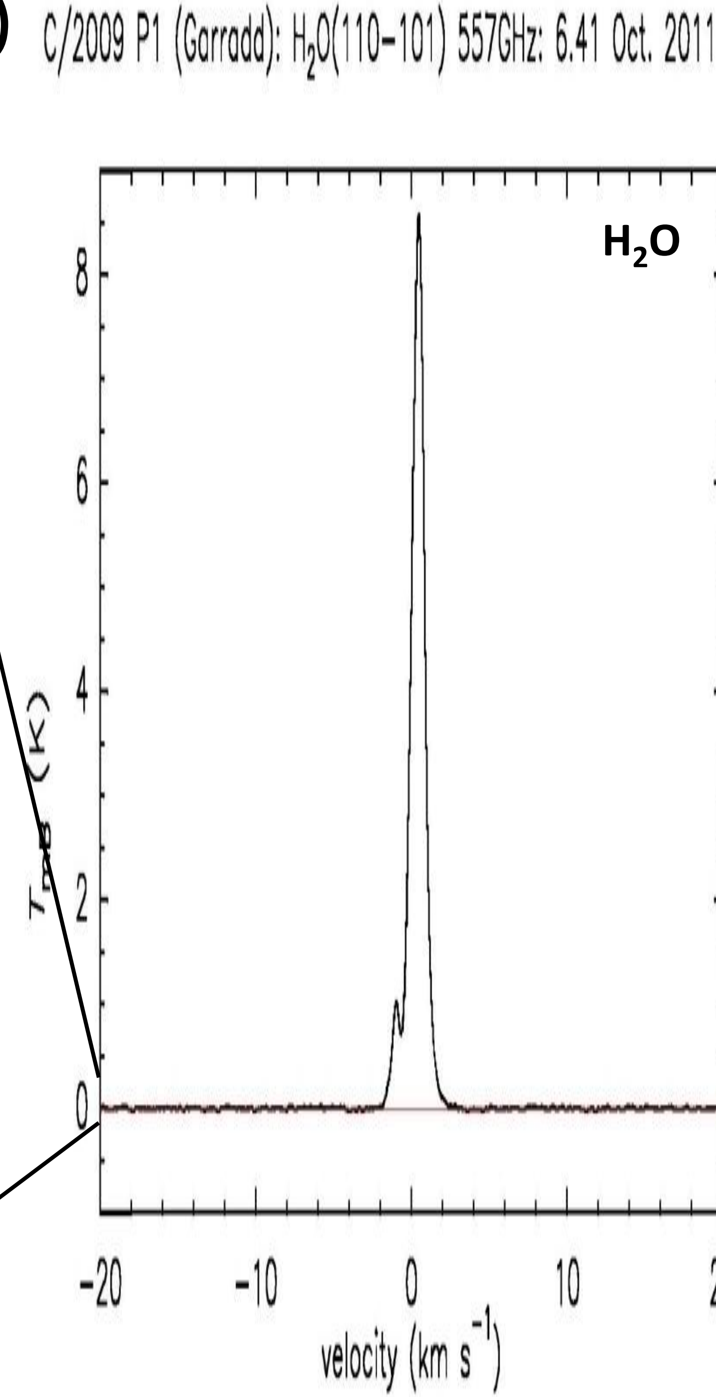
Line intensities ratio are: 1:8: 415

Production rates ratios are: 1:5:2430

($H_2^{16}O$ optically thick)





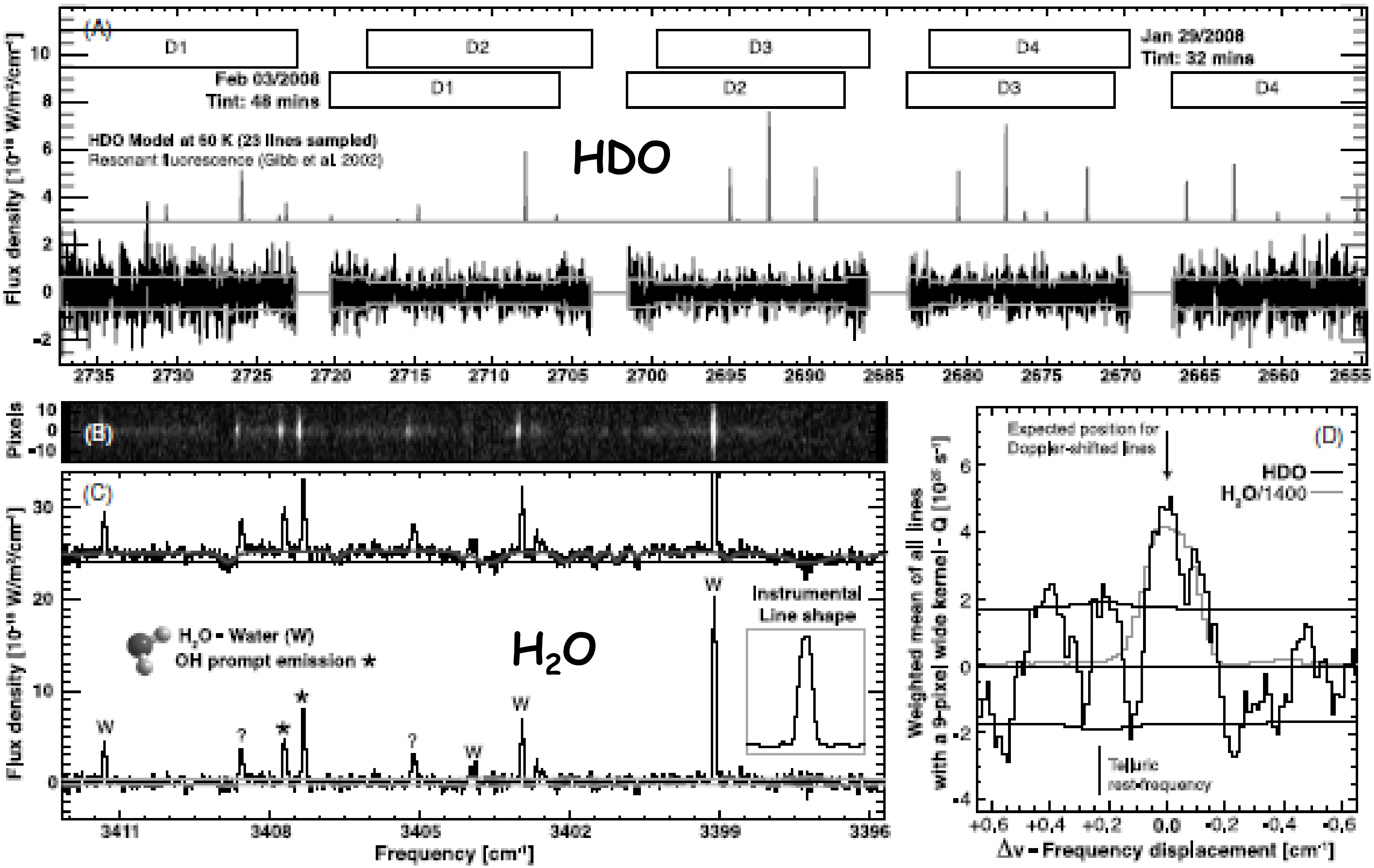


Bockelée-Morvan et al. 2012

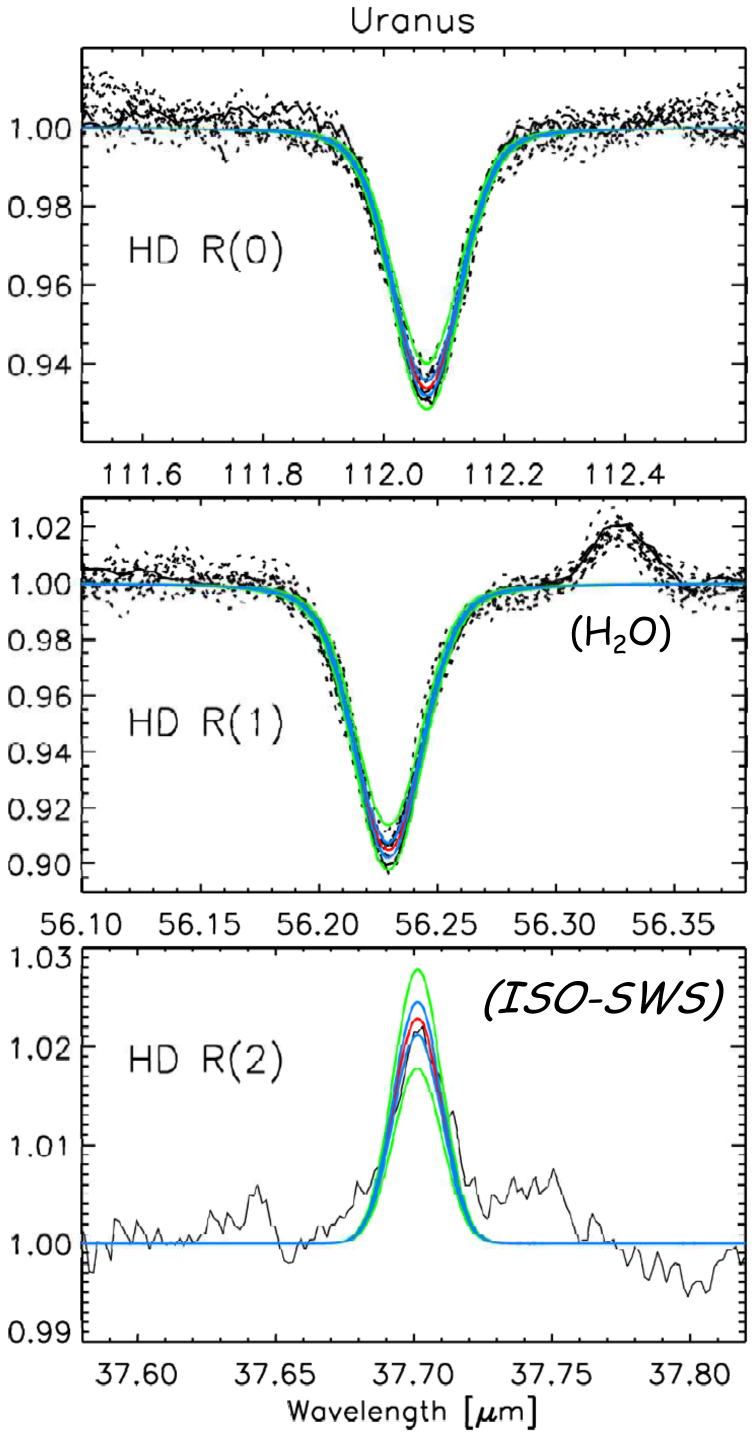
IR detection of HDO in comet 8P/Tuttle

(VLT-CRIRES Jan.-Feb. 2008 $\lambda/\delta\lambda$=42000)

Villanueva et al. 2009, ApJ 690 L5



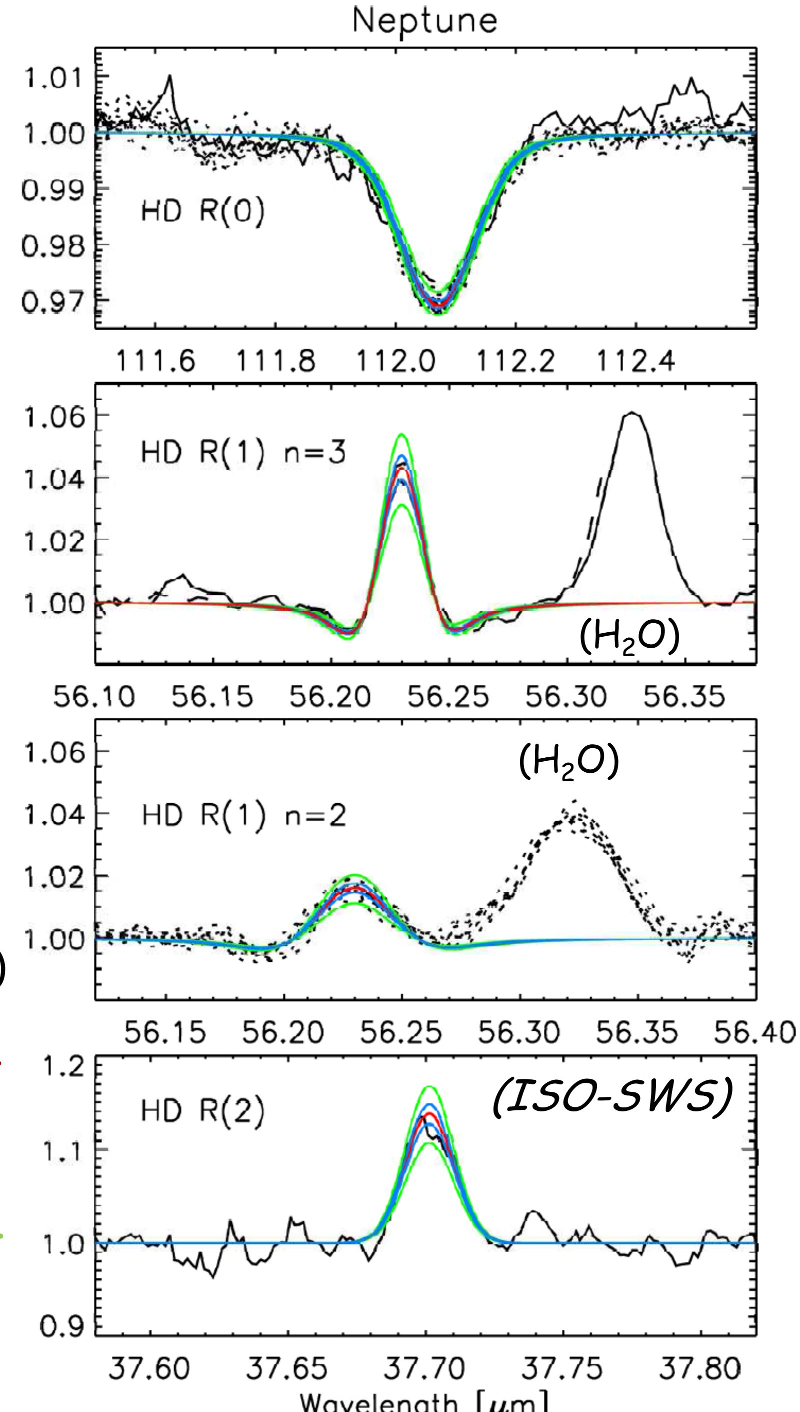
Average of 23 HDO lines



Herschel PACS observations of the HD R(0) and R(1) lines in Uranus and Neptune atmospheres.

(Feuchtgruber et al. 2013, A&A 551, A126)

Red = D/H best fit model,
blue: D/H ±1σ,
green: D/H ±3σ.



Isotopic ratios in Giant planets atmospheres:

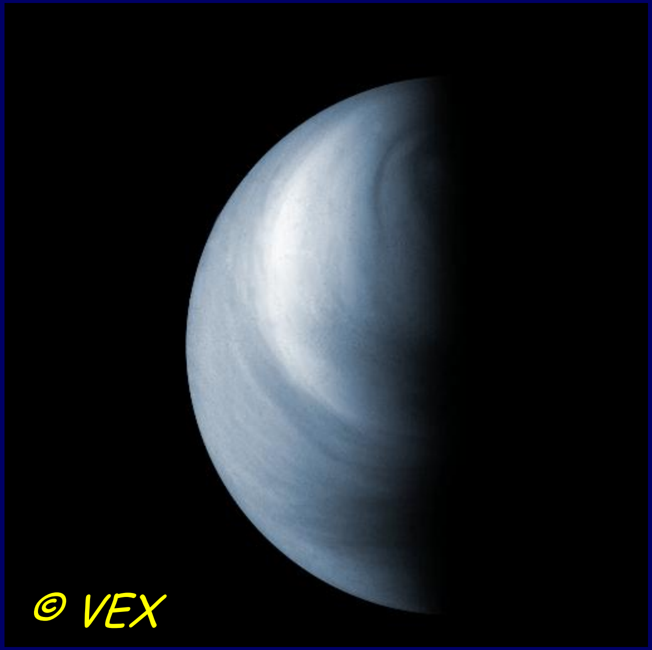
ratio	molecule	technique	value	planet	Ref.
D/H	H_2	HD, CH_3D, D (IR, MS Galileo)	**0.225**±0.035 ×10^{-4}	Jupiter	
		From HD and CH_3D (ISO)	**0.17**$(^{+0.08}_{-0.05})$×10^{-4}	Saturn	Griffin et al. 1996, Lellouch et al. 2001
		From CH_3D (Cassini/CIRS)	**0.16**±0.02 ×10^{-4}		Feltcher et al. 2009
		From HD submm (Herschel)	0.44±0.04 ×10^{-4}	Uranus	Feuchtgruber et al. 2013
		"	0.41±0.04 ×10^{-4}	Neptune	"
$^{14}N/^{15}N$	N	MS, Galileo	**435±60**	Jupiter	Owen et al. 2001
	NH_3	IR (IRTF)	**400-700**		Fletcher et al. 2014
	NH_3	IR (IRTF)	**>360**	Saturn	"

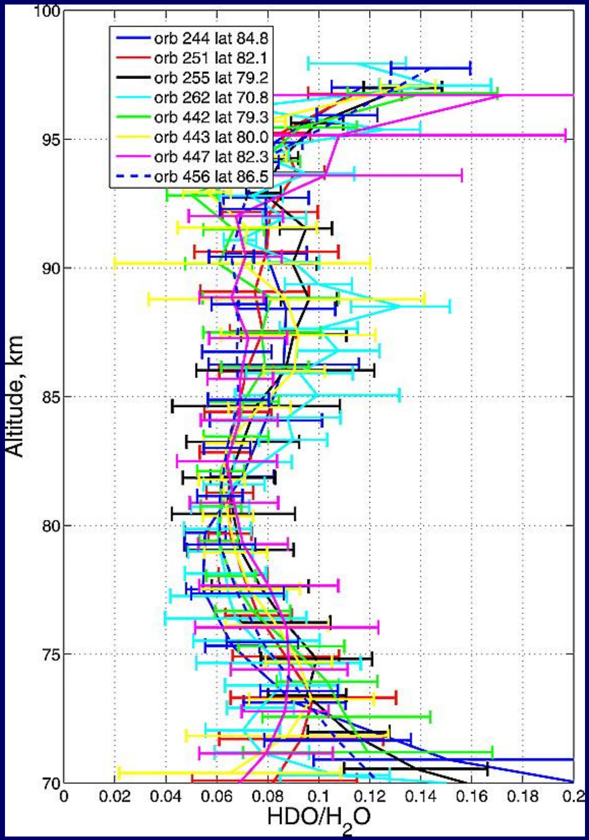
Telluric Planets Atmospheres: Venus

Venus Express SPICAV/SOIR (IR):

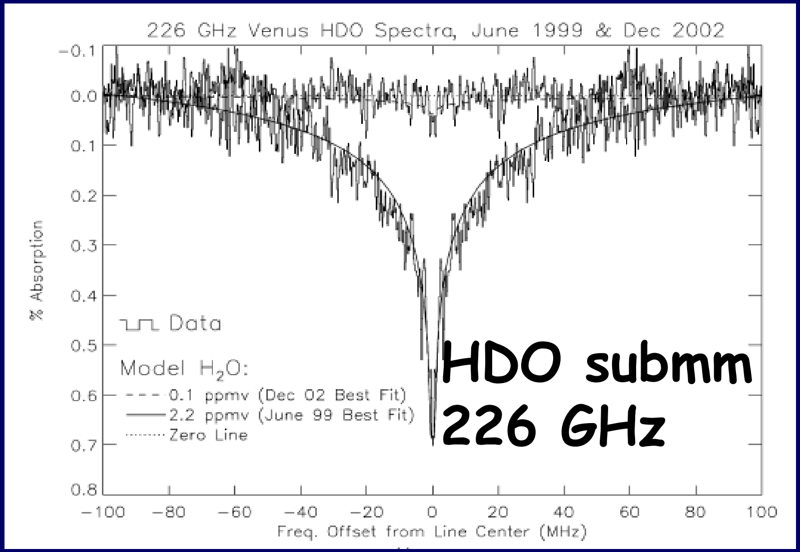
D/H in water vapour = 370±40 ×10^{-4}

(240×⊕) (Fedorova et al. 2008, JGR 113-E5, E00B22)



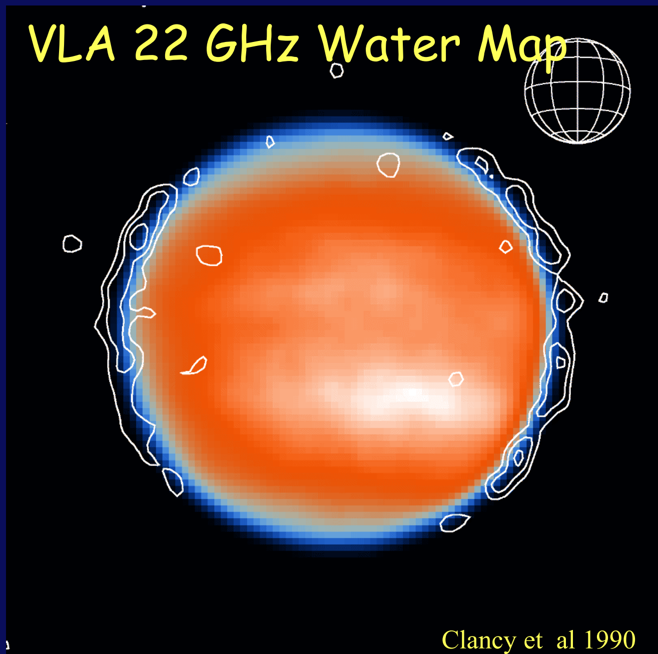


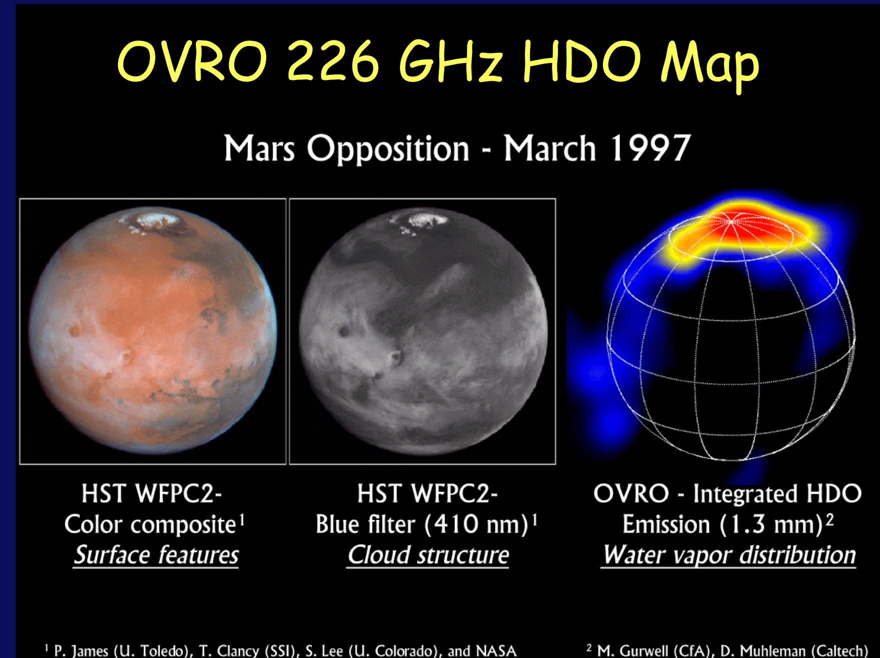
+ Huge variability of H_2O above Venus clouds

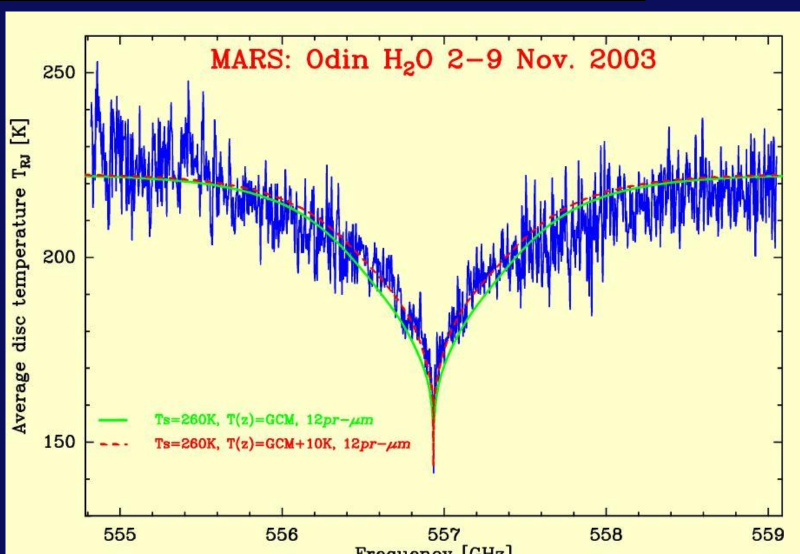


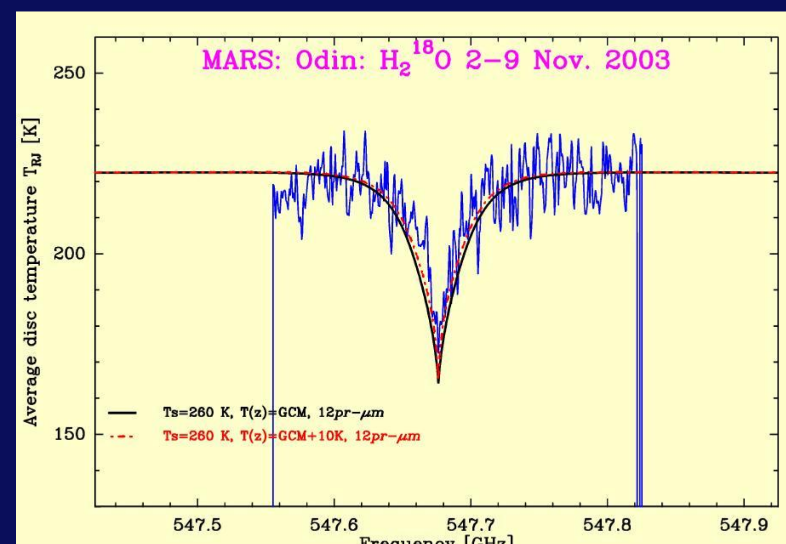
$^{14}N/^{15}N$ = 272±54 (MS, Pioneer Venus, Hoffman et al. 1979, Science, 205, 49)

Telluric Planets Atmospheres: Mars Water







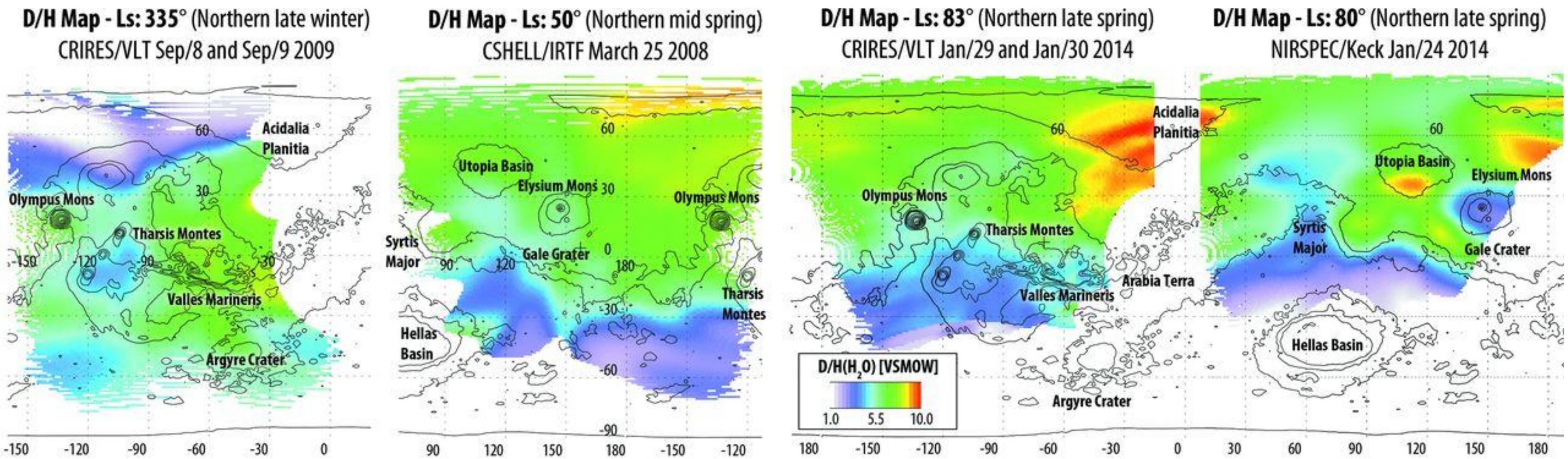


$^{16}O/^{18}O$ ~ 500 (terrestrial), (MS Viking, Owen et al. 1977, JGR 82, 4635)

Mars isotopic ratios

D/H in water: **9±4 ×10^{-4}** (IR, Owen et al. 1988, Science 240, 1767)

2-12×10^{-4} (IR: VLT/Keck/IRTF, Villanueva et al. 2015, Science 348, 218): local/seasonal variations of the D/H ratio



D/H in 3Gyr old clays: **4.7±0.3×10^{-4}** (MS: Curiosity/SAM, Mahaffy et al. 2015, Science 347, 412)

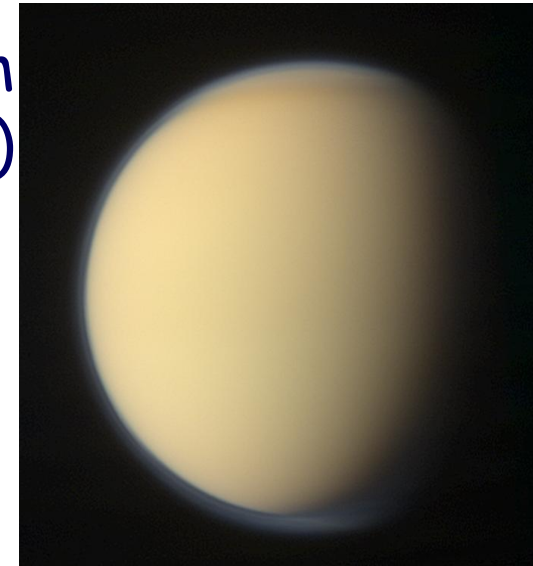
$^{14}N/^{15}N$ in N_2 = **173±11** (MS: Curiosity/SAM, Wong et al. 2013, GRL 40, 6033)

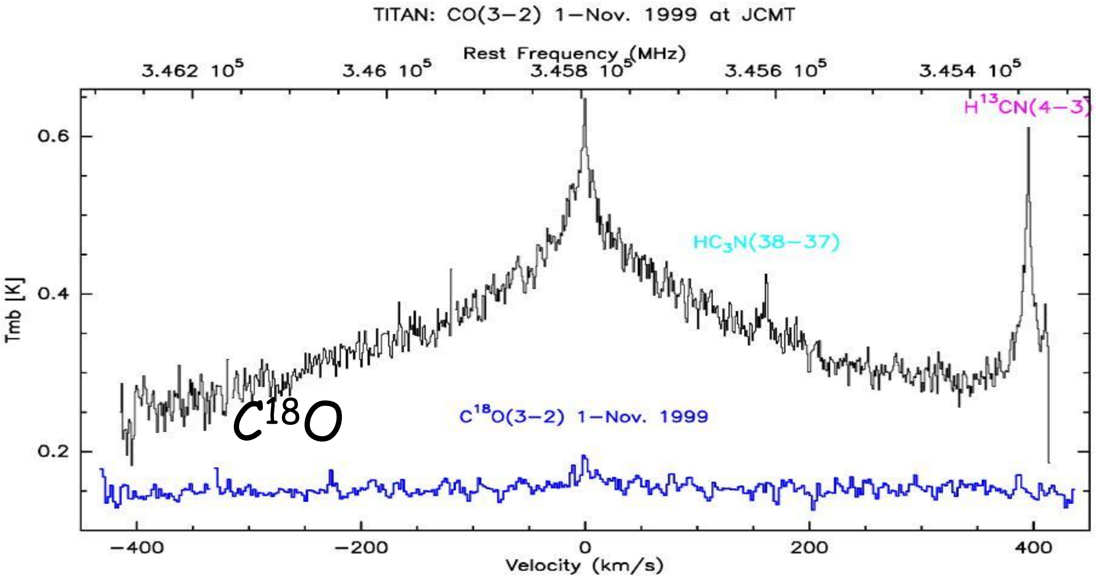
$^{12}C/^{13}C$ in CO_2 = **85±1** (MS: Curiosity/SAM, Mahaffy et al. 2013, Science 341, 263)

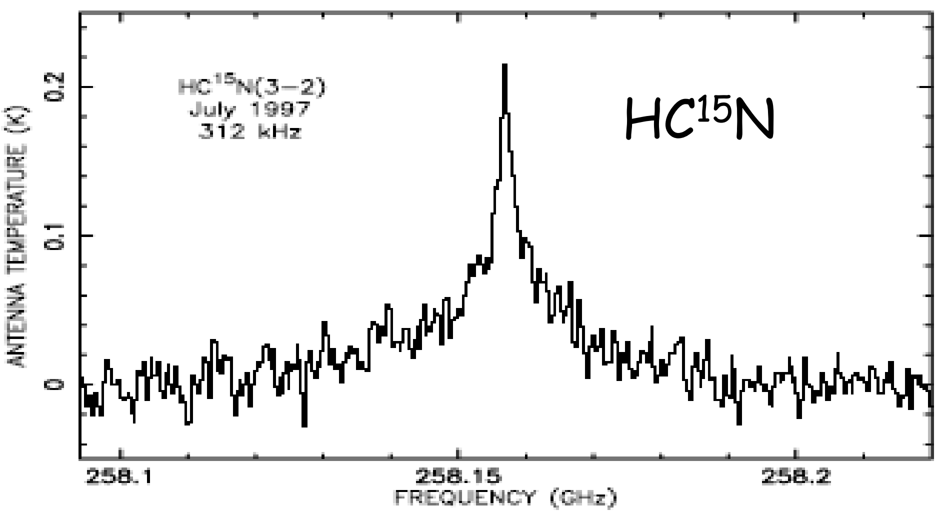
$^{36}Ar/^{40}Ar$ = **0.53±0.08 ×10^{-3}** (Earth: 3.4×10^{-3})

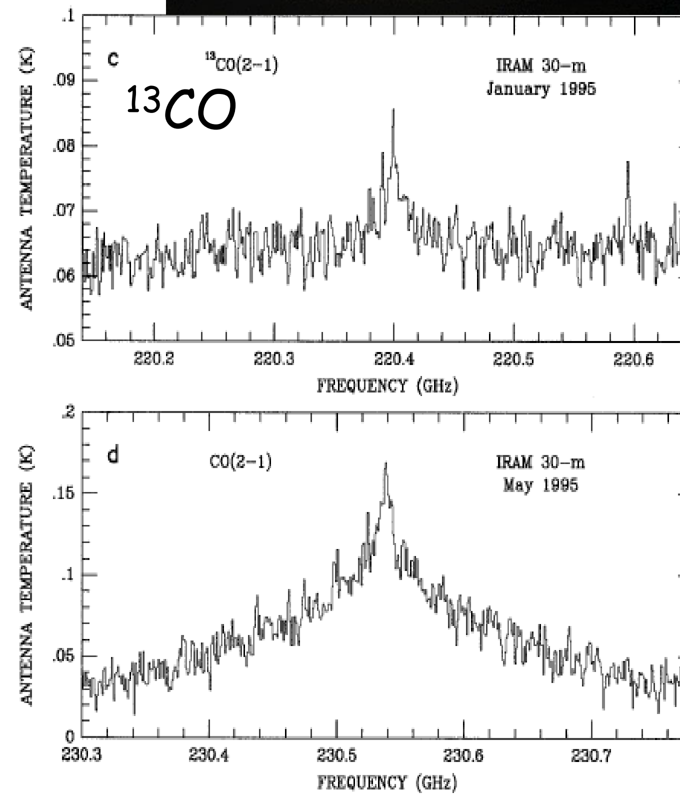
-> enrichment in D, ^{15}N, ^{13}C with time: atmospheric escape and soil<-> atmosphere exchanges

Titan: HCN, CO isotopes from the submm (IRAM, JCMT)





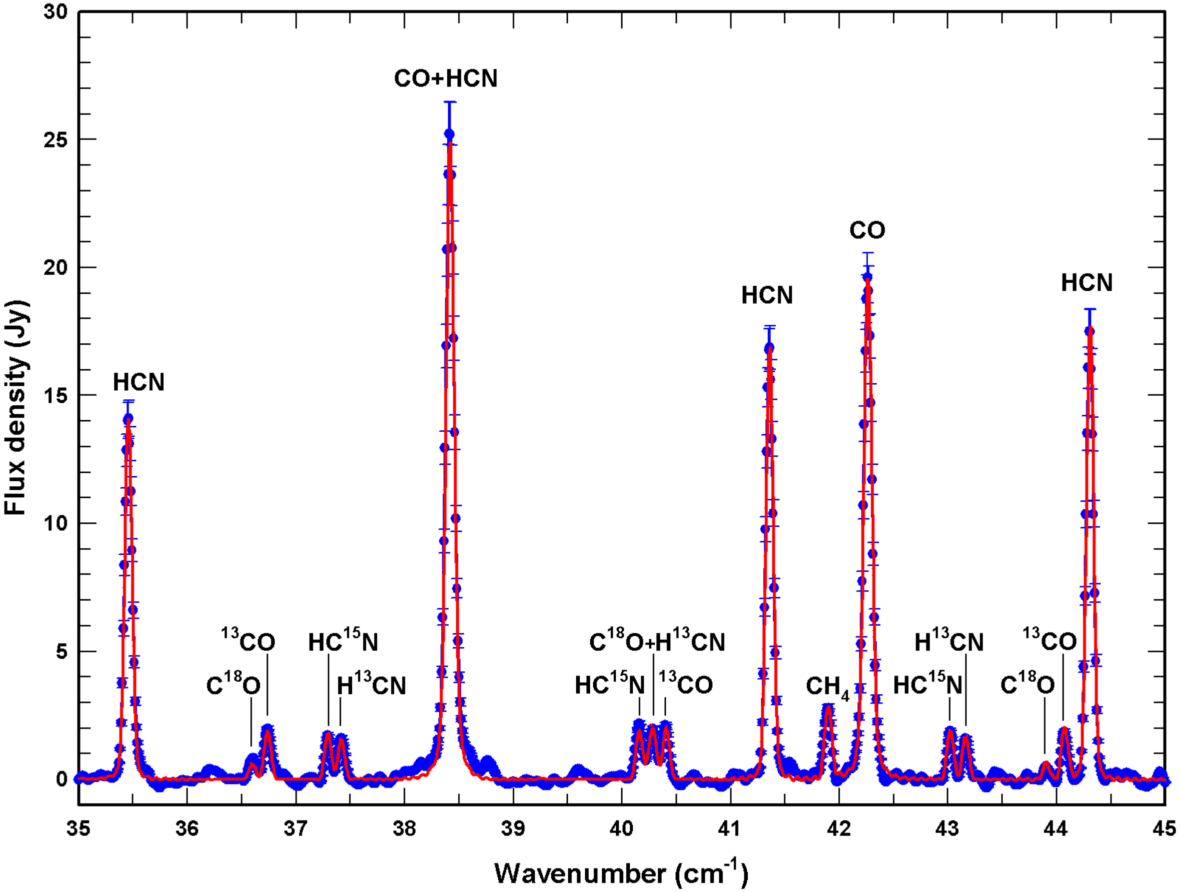




$^{15}N/^{14}N$: enriched 4.5 times compared to terrestrial value.

(Hydayat et al. 1998,…)

Titan: HCN, CO isotopes from the submm (Herschel-Spire)



Courtin et al. 2011

$^{16}O/^{18}O$ (in CO) = 380 ± 60
In CO_2 : 346 ± 111 (Nixon et al. 2008)
Anomaly in Oxygen source ? (Enceladus torus)

$^{12}C/^{13}C$ =87±6 (in CO)
96±13 (in HCN)
Consistent with previous measurements in submm and from Cassini

$^{14}N/^{15}N$ = 76±6 (in HCN)
Huygens-GCMS: **183±5 in N_2**

=> Photolytic fractionation of N_2 leading to HCN enrichment in ^{15}N (Liang et al. 2007)

Isotopic ratios in Titan atmosphere *(blue=Earth, green: 1-2x, red:3-4x)*

From Bézard et al. 2014, in « Titan, interior, Surface, Atmosphere and Space Environment », Eds I. Müller-Wodarg, C.A. Griffith, E. Lellouch and T. Cravens - Cambridge Planetary Science

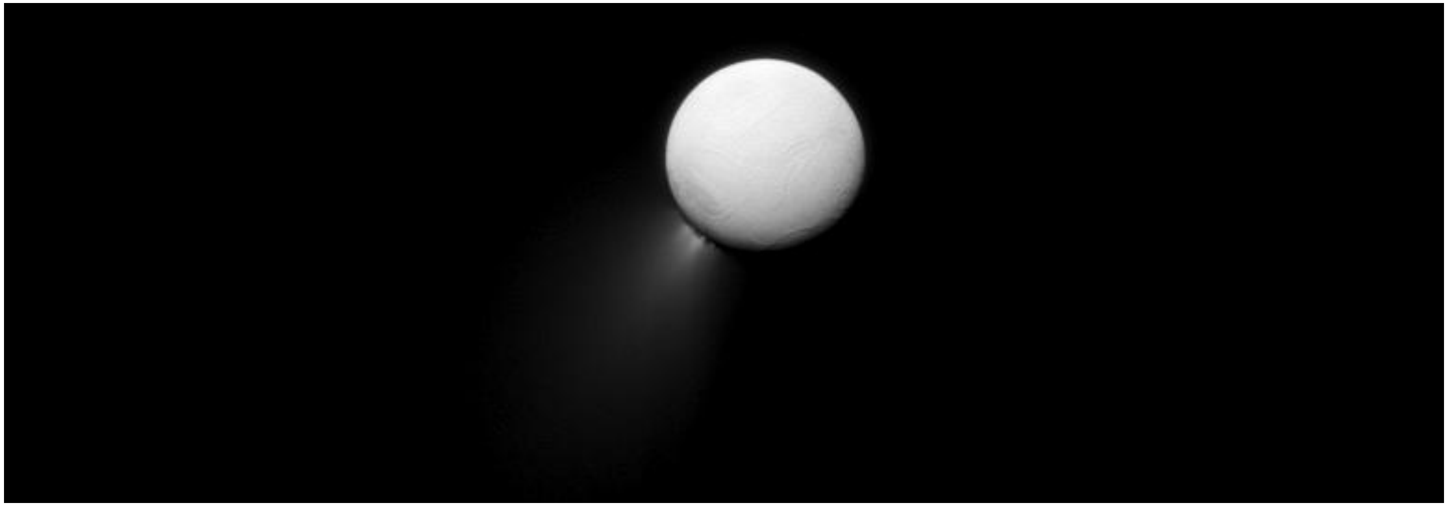
Ratio	molecule	technique	value	Altitude	Ref.
D/H	H_2	GCMS, Huygens	$1.4\pm0.3\times10^{-4}$	0-120 km	Niemann et al. 2010
	CH_4	Near-IR,IRTF, KPNO	$1.1\pm0.3\times10^{-4}$	0-50 km	de Bergh et al. 2012
		IR - IRTF	$1.3\pm0.3\times10^{-4}$	95-290 km	Penteado et al. 2005
		IR/CIRS-Cassini	$1.2\text{-}1.6\times10^{-4}$	75-290 km	several
	C_2H_2	IR/CIRS-Cassini	$1.6\text{-}2.1\times10^{-4}$	115 km	Coustenis et al. 2008
$^{14}N/^{15}N$	N_2	GCMS, Huygens	167±1	16-144 km	Niemann et al. 2010
		INMS, Cassini	168-211	(950=>)0km	Waite et al. 2005
	HCN	submm (IRAM, SMA)	60-72	80-400 km	Marten et al. 2002, Gurwell 2004, 2011
		submm (Herschel)	76±6	90-230 km	Courtin et al. 2011
		IR/CIRS Cassini	56±8	165-305 km	Vinatier et al. 2007

Isotopic ratios in Titan atmosphere

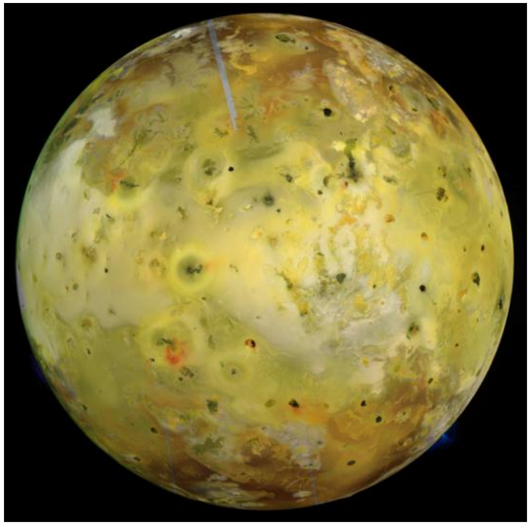
Ratio	molecule	technique	value	Altitude	Ref.
$^{12}C/^{13}C$	CH_4	GCMS, Huygens	91±1	0-120 km	Niemann et al. 2010
		IR/CIRS-Cassini	86-89	0-275 km	Mandt et al. 2012, Nixon et al. 2012
	C_2H_2,C_2H_6, C_4H_2	IR/CIRS-Cassini	85-90	100-150 km	Nixon et al. 2012, Jolly et al. 2010
	HCN	IR/CIRS-Cassini	75±12	165-305 km	Vinatier et al . 2007
		Submm (Herschel,SMA)	96-132	80-400 km	Courtin et al. 2011, Gurwell 2004
	HC_3N	IR/CIRS-Cassini	79±17	150 km	Jennings et al . 2008
	CO	Submm (Herschel,SMA)	84-87	60-230 km	Courtin et al. 2011, Gurwell et al. 2011
	CO_2	IR/CIRS-Cassini	84±17	100-150km	Nixon et al. 2008
$^{16}O/^{18}O$	CO	submm (JCMT, SMA, Herschel)	250-472	60-300 km	Courtin et al. 2011, Gurwell et al. 2011, Owen et al. 1999
	CO_2	IR/CIRS-Cassini	346±111	100-150km	Nixon et al. 2008
$^{36}Ar/^{40}Ar$	Ar	GCMS, Huygens	$6.1\pm2.5 \times 10^{-3}$	75-77 km	Niemann et al. 2010

Isotopic ratios in other minor bodies:

Enceladus water: D/H = $2.9(^{+1.5}_{-0.7}) \times 10^{-4}$ (Waite et al. 2009)



Sulfur isotopes in **Io**: $^{32}S/^{34}S$ in SO_2 = 8-15

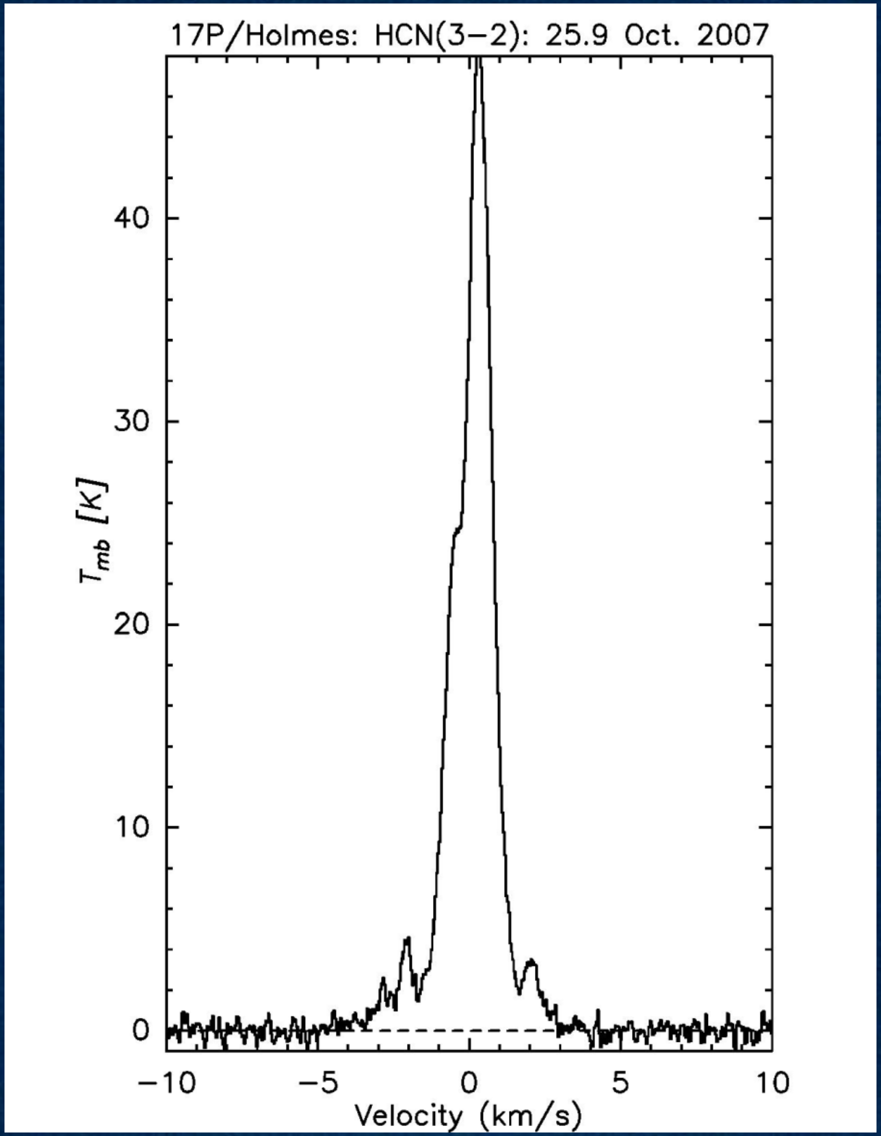


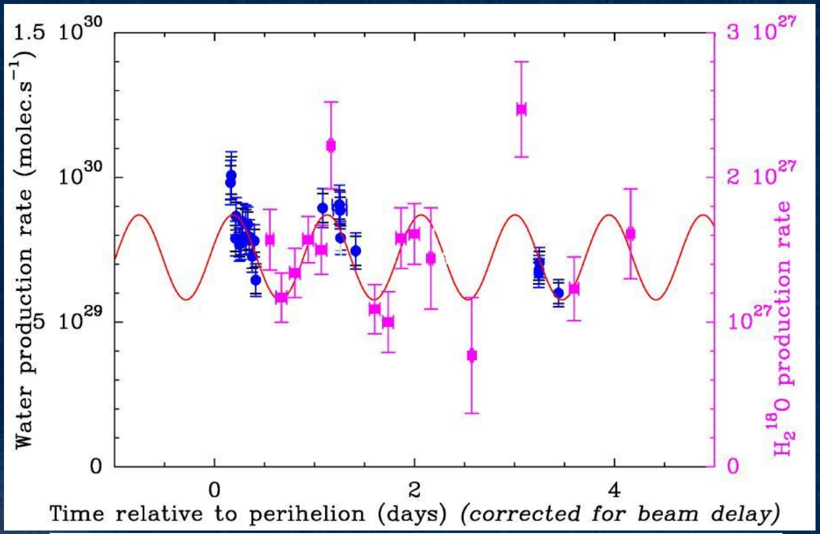
(SO_2 gas with APEX (submm):
Moullet et al. 2013 ApJ 776, 32)

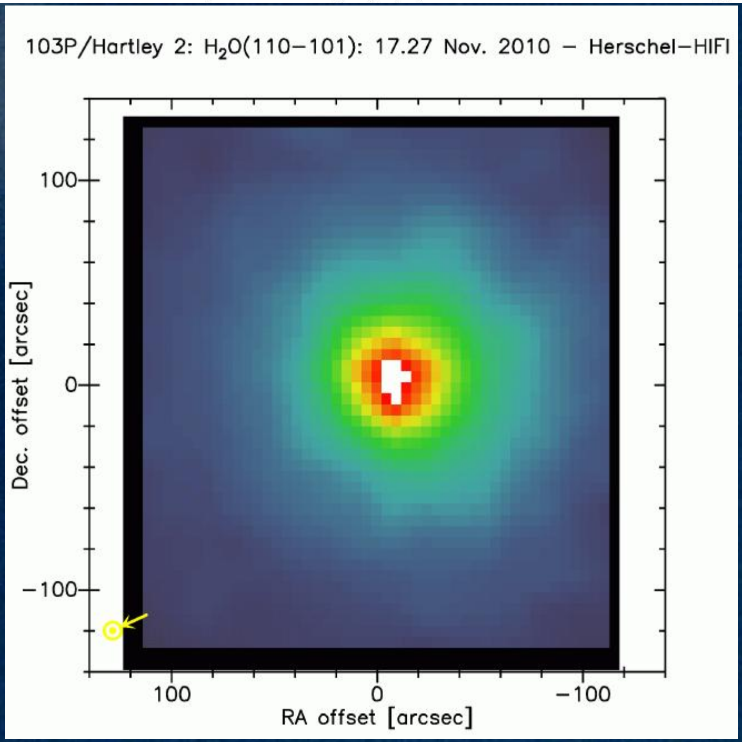
$^{34}S/^{33}S$ = 7.7±4.1

(SO_2 frost with UKIRT (IR):
Howell et al. 1989, Icarus 78, 27)

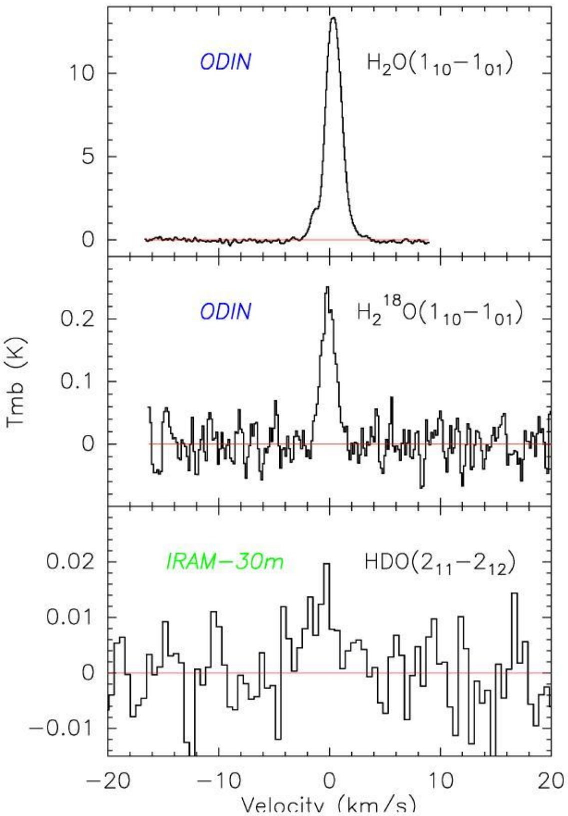
Measuring Isotopic ratios in comets - time variability:

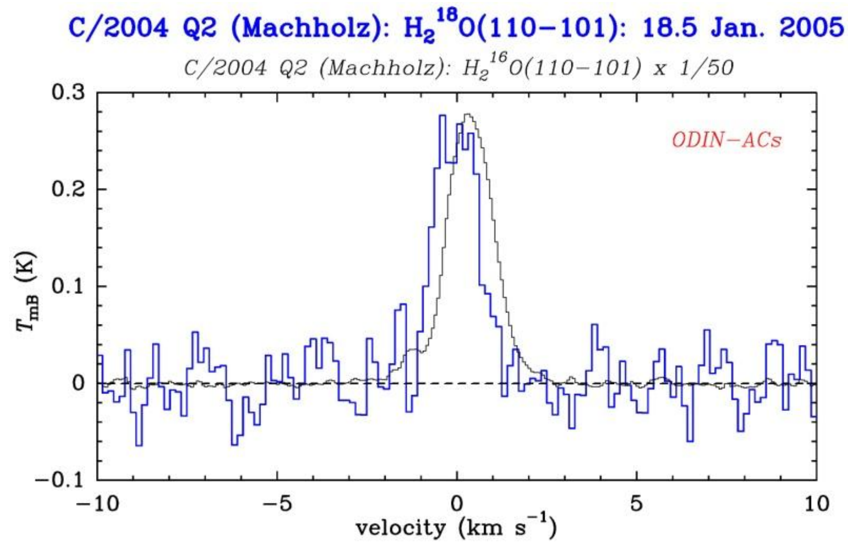


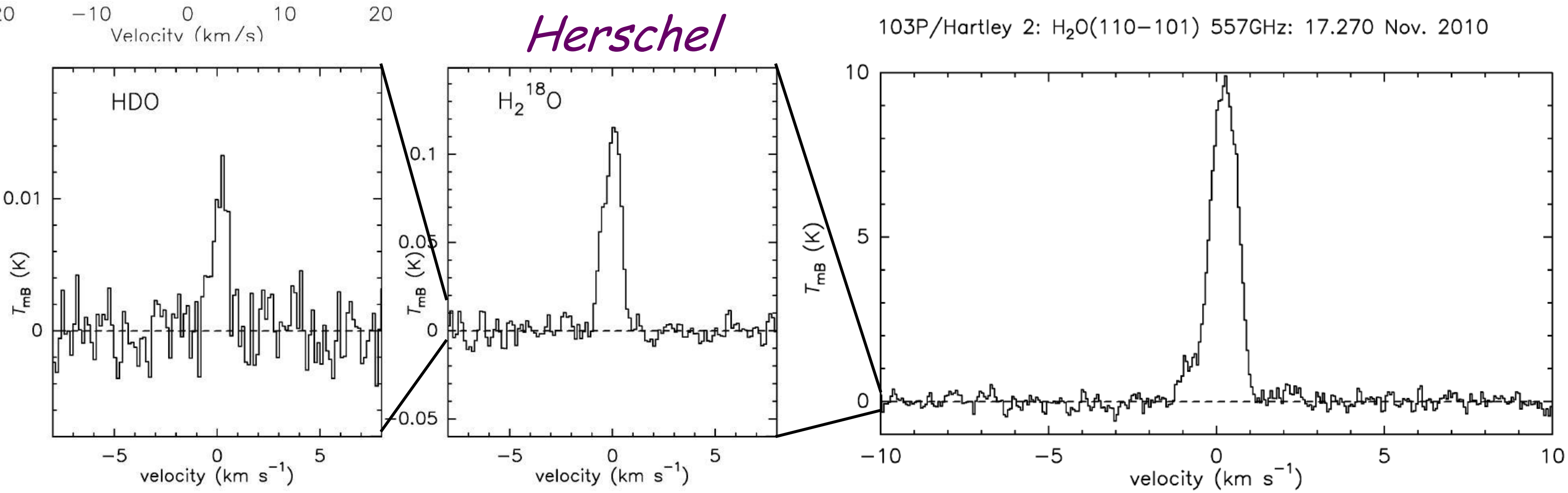




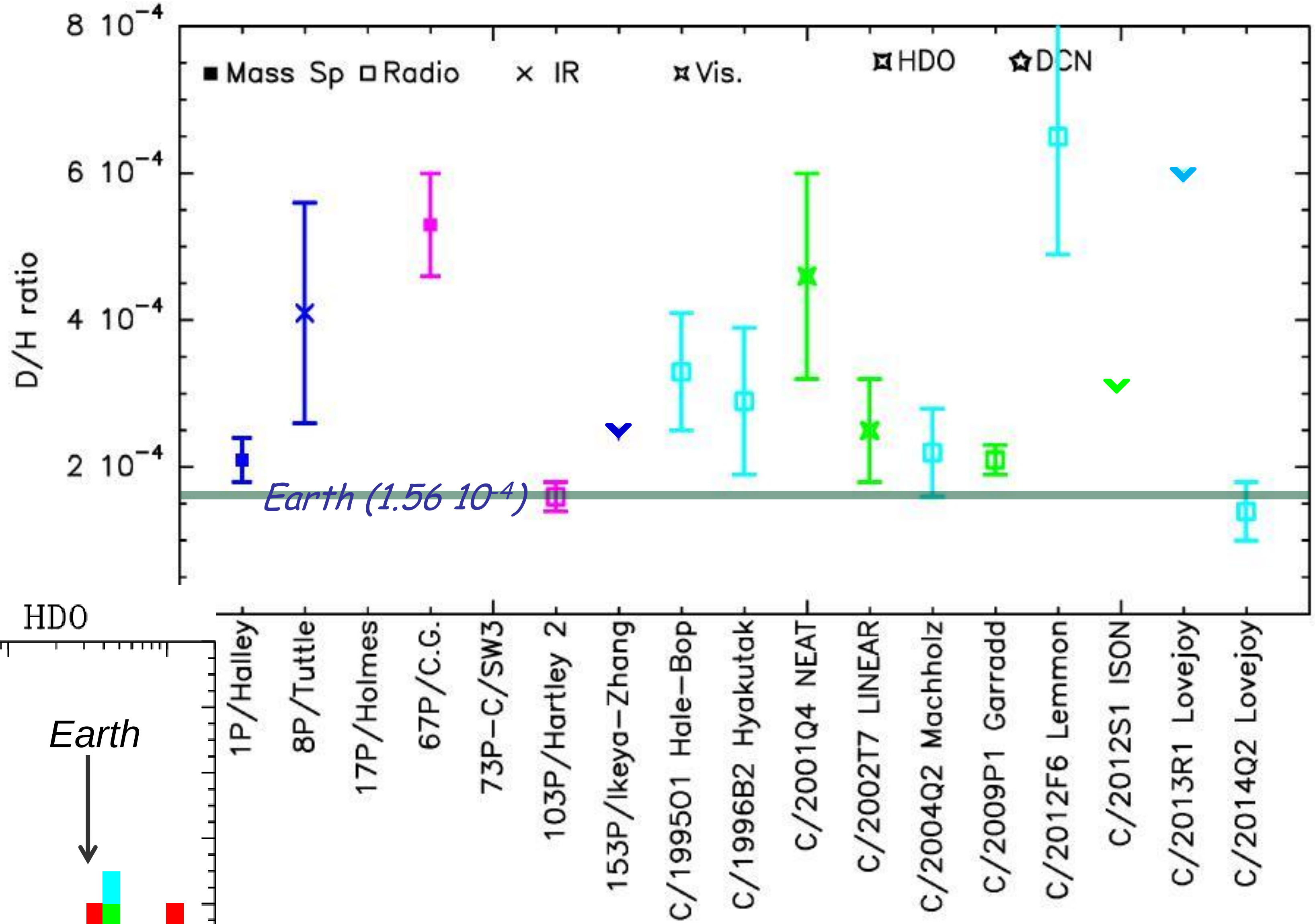
Water Isotopologues in comets

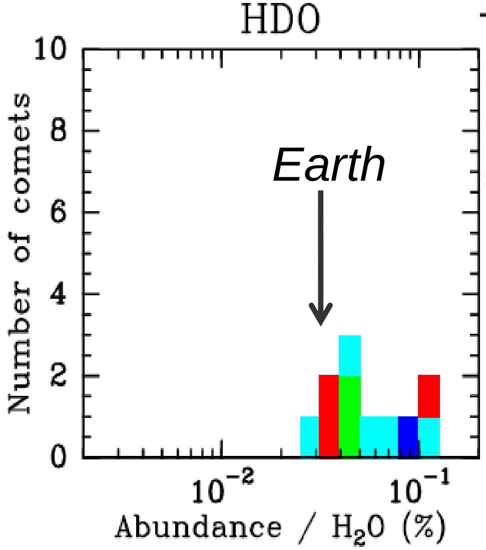






D/H measurements in cometary water:





D/H ratio in cometary water

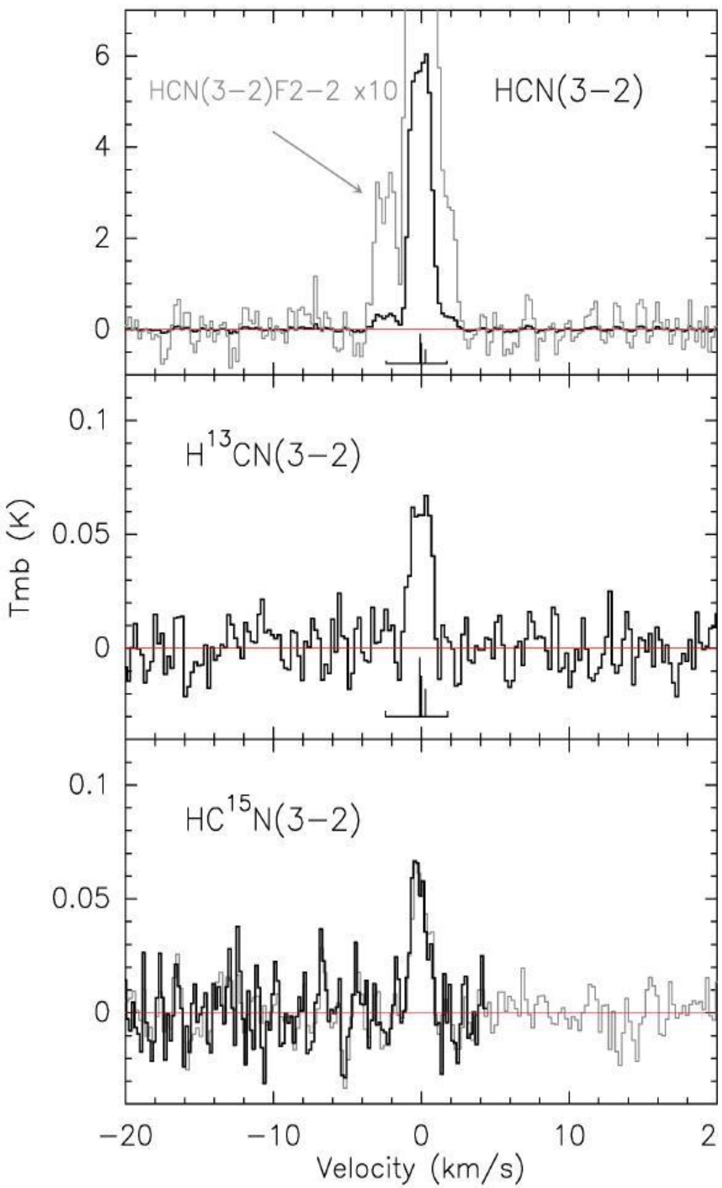
Obs. species	technique	Comet (JFC,HFC,LP,DN)	D/H value (×10^{-4})	Reference
H_2DO^+	Ion mass spect.	1P/Halley	2.1 ±0.3	Brown et al. 2012
HDO	radio	C/1996 B2 (Hyakutake)	2.9±1.0	Bockelée-Morvan et al. 1998
	radio	C/1995 O1 (Hale-Bopp)	3.3±0.8	Meier et al. 1998
	IR	8P/Tuttle	4.1±1.5	Villanueva et al. 2009
	radio	103P/Hartley 2	1.6±0.2	Hartogh et al. 2011
	radio	C/2009 P1 (Garradd)	2.1±0.2	Bockelée-M. et al. 2012
	radio	45P/H.M.P.	< 2.0	Lis et al. 2013
	radio	153P/Ikeya-Zhang	< 2.5	Biver et al. 2006
	radio	C/2004 Q2 (Machholz)	2.2±0.6	*Biver in prep.*
	Mass spectro.	67P/Churyumov-G.	5.3±0.7	Altwegg et al. 2015
	radio	C/2014 Q2 (Lovejoy)	1.4±0.4	Biver et al. 2016
	radio	C/2012 F6 (Lemmon)	6.5±1.6	Biver et al. 2016
	IR	C/2012 S1 (ISON)	< 3.1	Gibbs et al. 2016
OD	uv-visible	C/2002 T7 (LINEAR)	2.5±0.7	Hutsemékers et al. 2008
D	uv	C/2001 Q4 (NEAT)	4.6±1.4	Weaver et al. 2008

D/H ratio in cometary molecules (best upper limits, remote)

Obs. species	technique	Comet	D/H value ($\times 10^{-4}$)	Reference
DCN	radio	C/1995 O1 (Hale-Bopp)	**23±5**	Meier et al. 1998b Crovisier et al. 2004
	radio	C/1996 B2 (Hyakutake)	*<100*	Bockelée-Morvan et al. 1998
	radio	C/2014 Q2 (Lovejoy)	*< 60*	Biver et al. 2016
HDCO	radio	C/2014 Q2 (Lovejoy)	*< 70*	Biver et al. 2016
HDS	radio	C/2014 Q2 (Lovejoy)	*<170*	Biver et al. 2016
	radio	17P/Holmes	*< 80*	Biver et al. 2008
CH_3D	IR	C/2004 Q2 (Machholz)	*< 50*	Bonev et al. 2009
	IR	C/2004 Q2 (Machholz)	*< 64*	Kawakita et al. 2009
	IR	C/2007 N3 (Lulin)	*< 75*	Gibbs et al. 2012
$ND(NH_3)$	visible	C/1996 B2 (Hyakutake)	*< 60*	Meier et al. 1998c

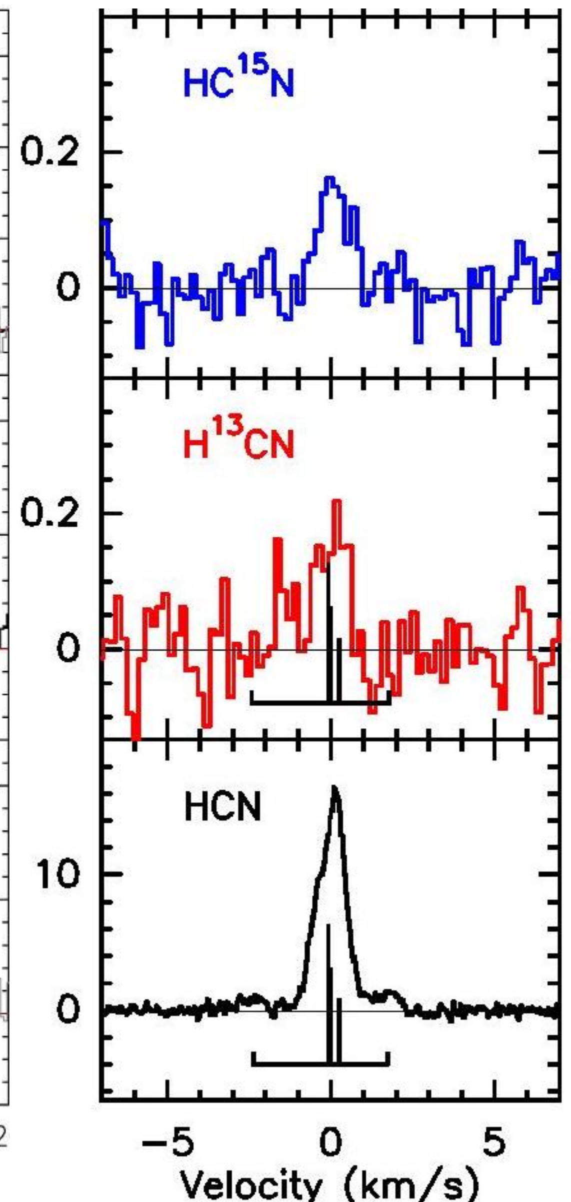
Nitrogen isotopic ratios

C/2014 Q2 (Lovejoy)



Biver et al. 2016

17P/Holmes



Bockelée-Morvan et al. 2008

C/2012 S1 (ISON)

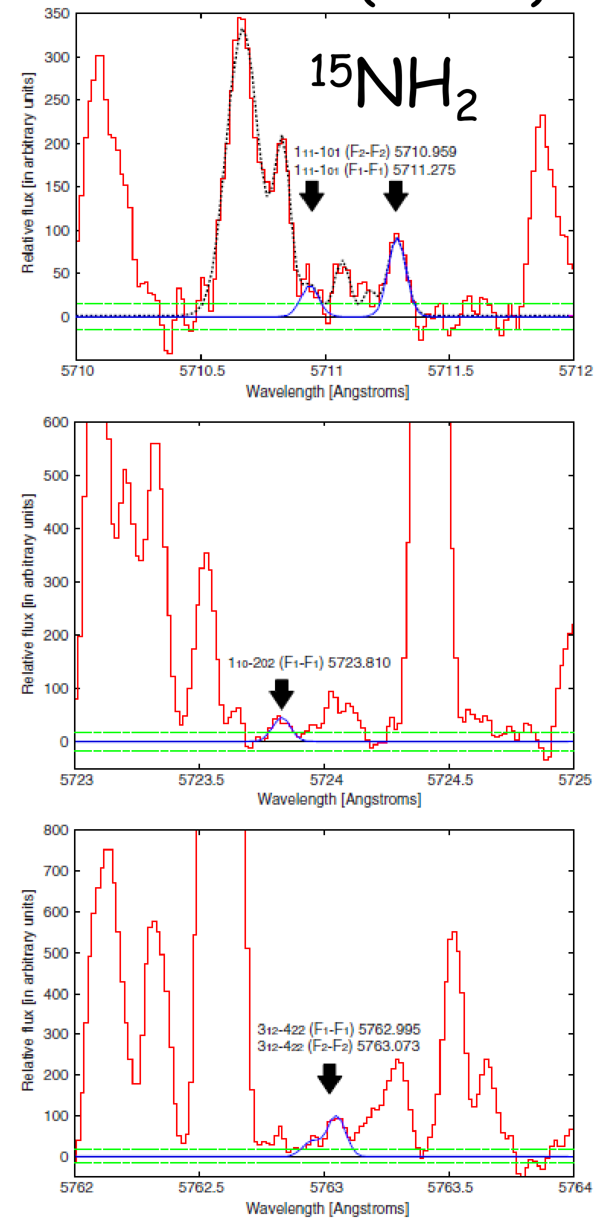
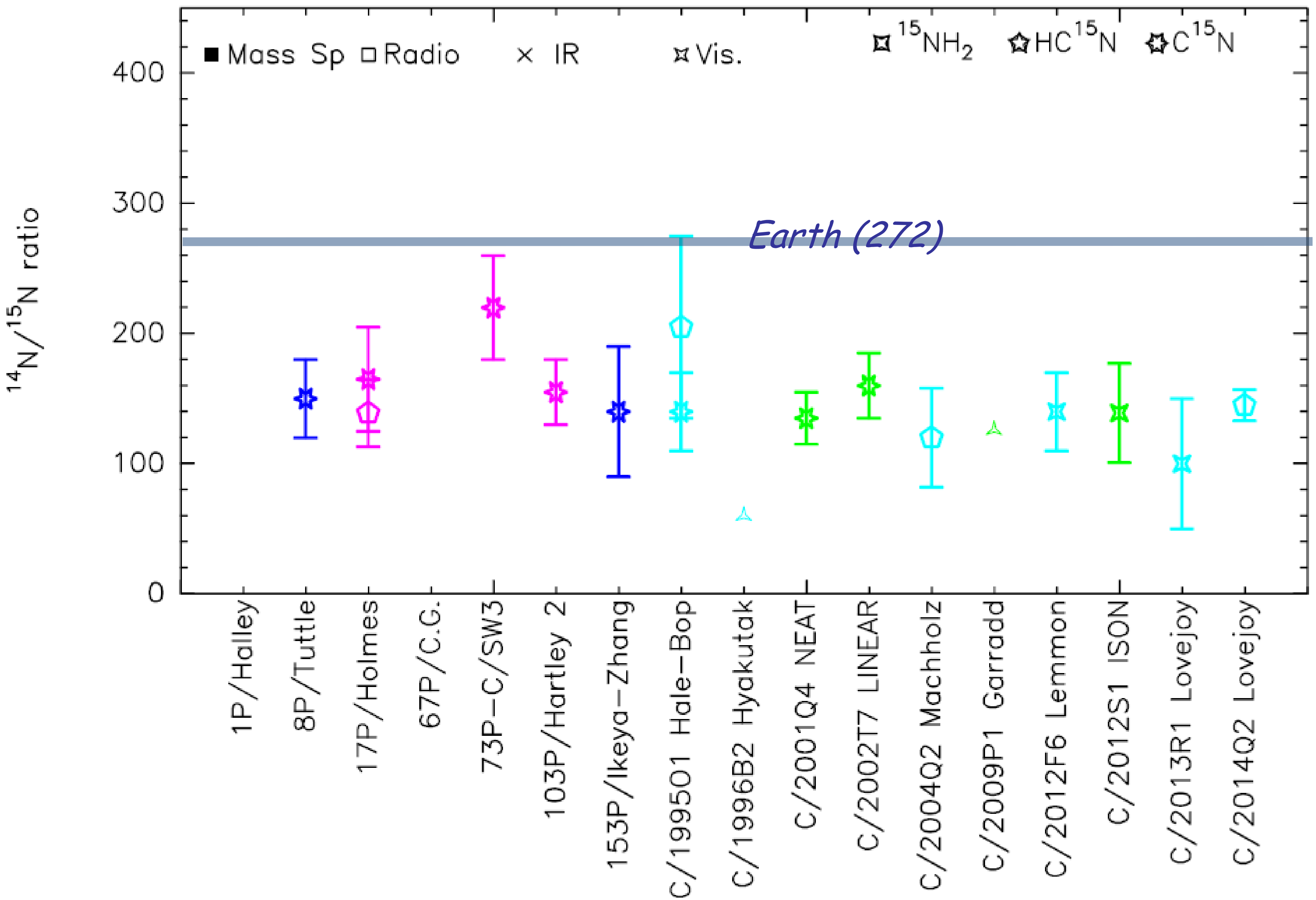


Figure 2. Three panels show the $^{15}NH_2$ emission lines detected in our spectrum.

Shinnaka et al. 2014

$^{14}N/^{15}N$ in comets



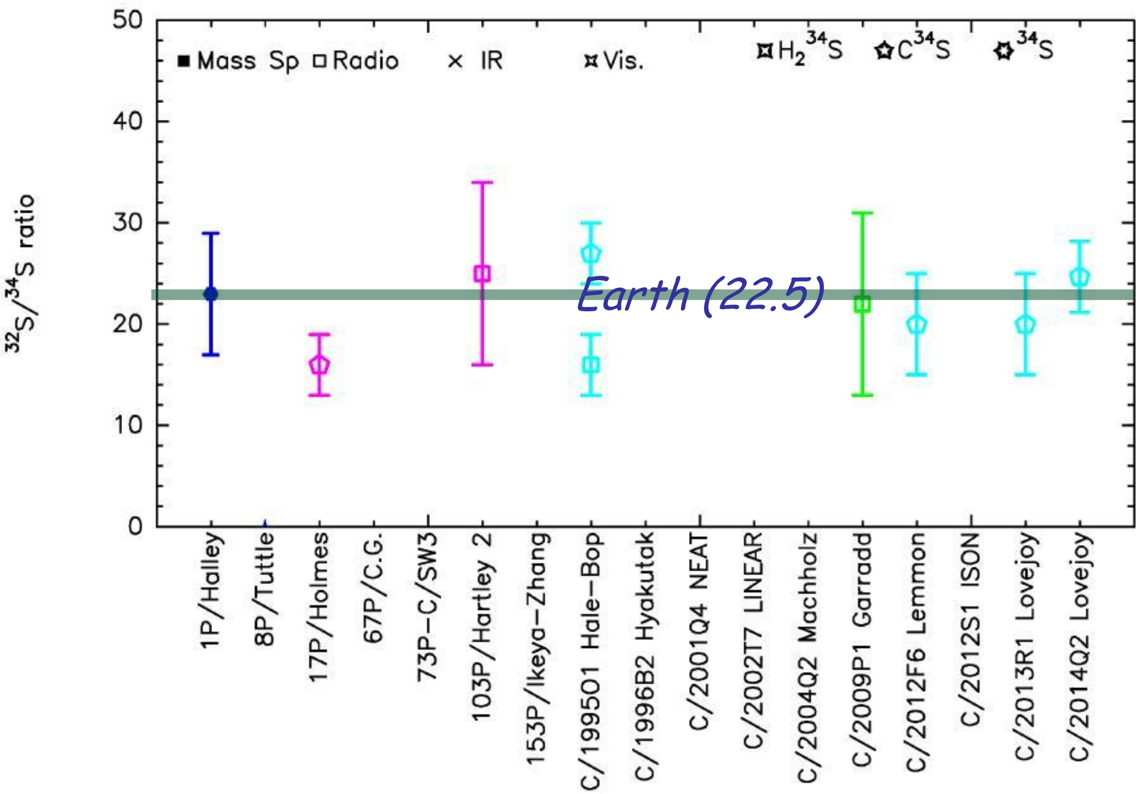
Nitrogen isotopic ratio in cometary molecules

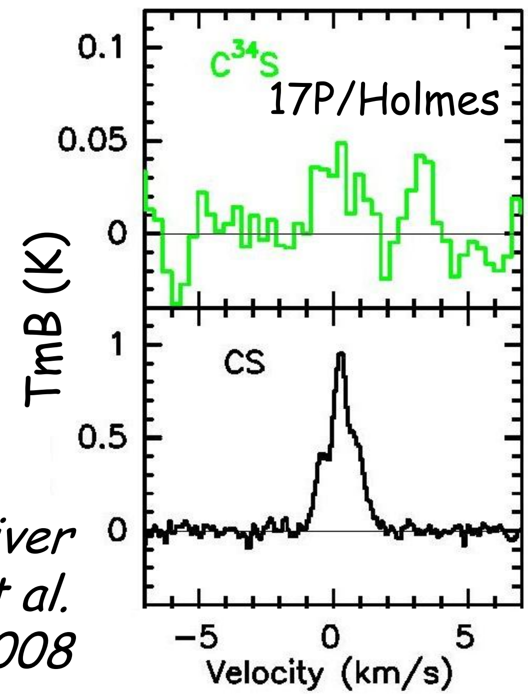
Ratio	Obs. species	techniq ue	Comet	ratio	Reference
$^{14}N/^{15}N$	HCN	radio	C/1995 O1 (Hale-Bopp)	205±70	Bockelée-Morvan et al. 2008
		radio	17P/Holmes	139±26	Bockelée-Morvan et al. 2008
		radio	C/2014 Q2 (Lovejoy)	145±12	Biver et al. 2016
	CN	visible	21 comets	141±29	Manfroid et al. 2009
		visible	73P-C/Schwassmann-W.3	220±40	Manfroid et al. 2009
		visible	17P/Holmes	165±40	Bockelée-Morvan et al. 2008
		visible	103P/Hartley 2	155±25	Jehin et al. 2011
	NH_2	visible	12 comets	90-190	Rousselot et al. 2014
		visible	C/2012 F6 (Lemon)	140±30	Decock et al. 2014
		visible	C/2012 S1 (ISON)	139±38	Shinnaka et al. 2014

Sulfur isotopes:

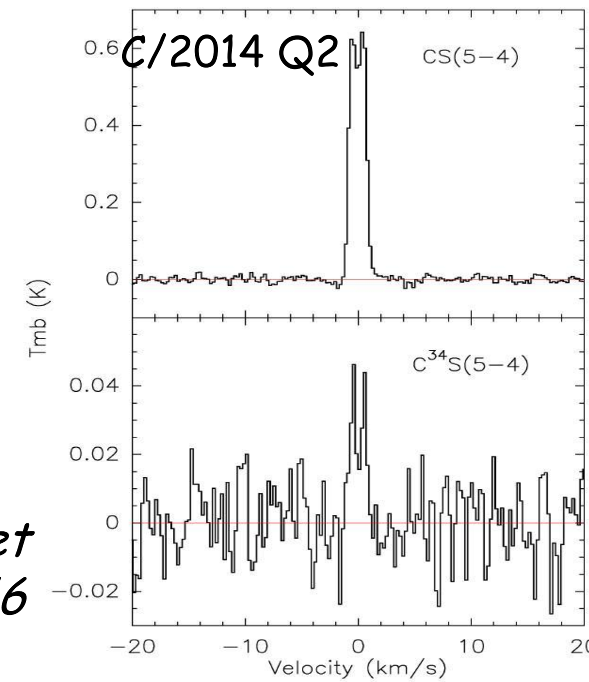
$H_2{}^{34}S$ and $C^{34}S$ (radio), ^{34}S, ^{33}S in-situ MS

$H_2{}^{33}S$ and $C^{33}S$ (Earth $^{32}S/^{33}S=127$) detectable in bright comets (e.g. $C^{32}S/C^{33}S > 50$ in C/2014 Q2)



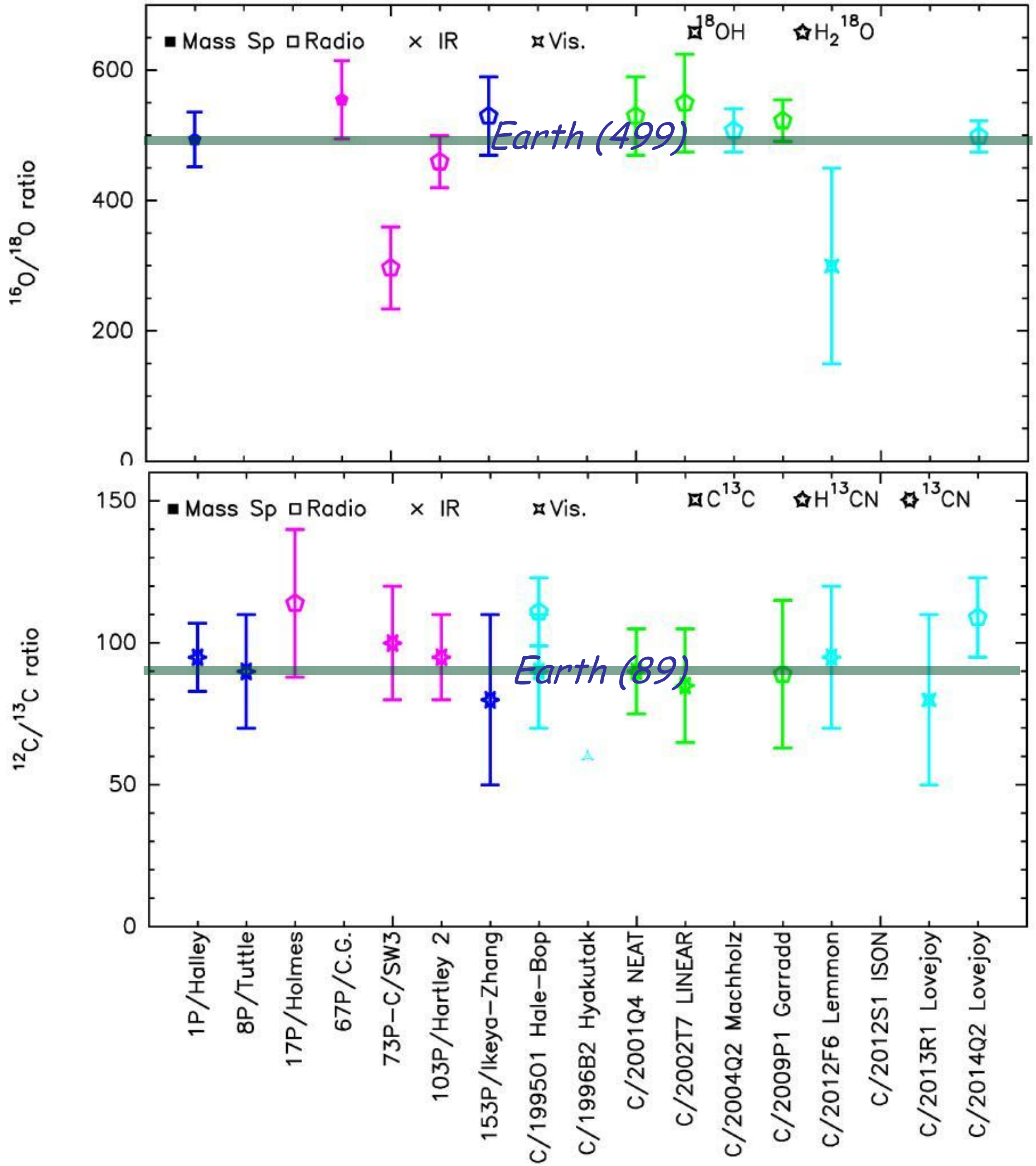


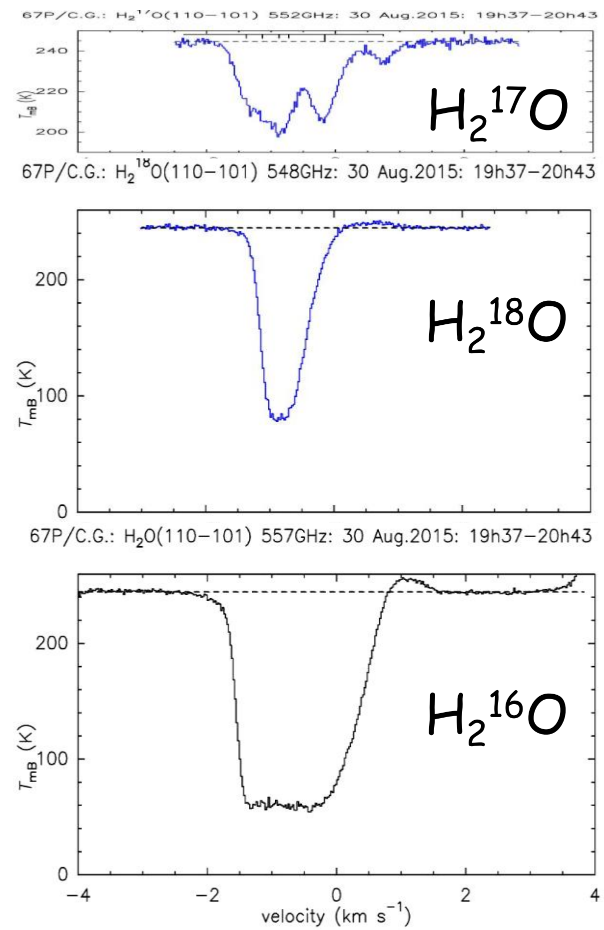
Biver et al. 2008



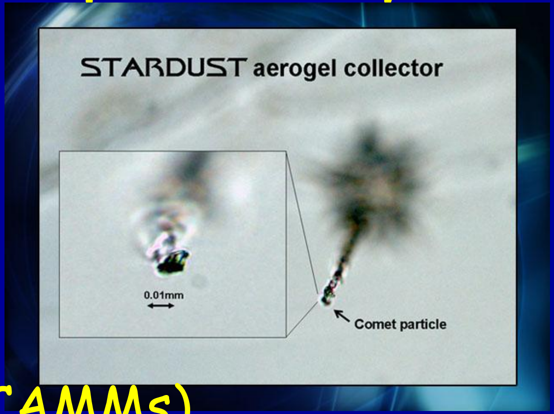
Biver et al. 2016

$H_2{}^{16}O/H_2{}^{17}O$: Only measured in-situ in comet 67P: Rosina, MIRO ~terrestrial (2682)





Isotopic ratio from extraterrestrial samples analysed in the laboratories



(1) Cometary sample: dust from 81P/Wild 2 collected by stardust (no volatiles)
(2) Interplanetary Dust Particles (IDPs) collected in the stratosphere (comet related meteoritic streams)
(3) Ultracarbonaceous Antarctic Micrometeorites (UCAMMs)
(4) Carbonaceous chondrites

(4) Robert 2013

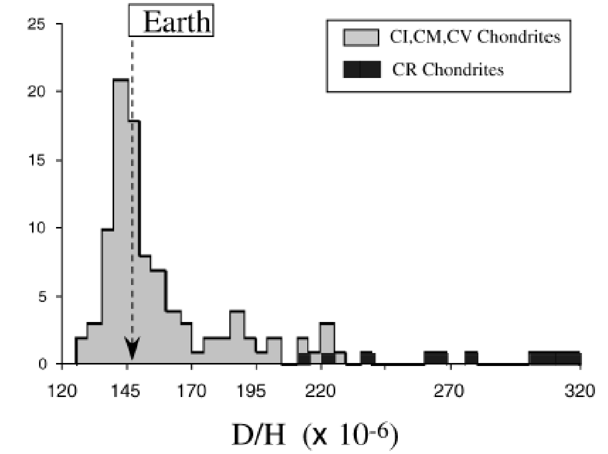
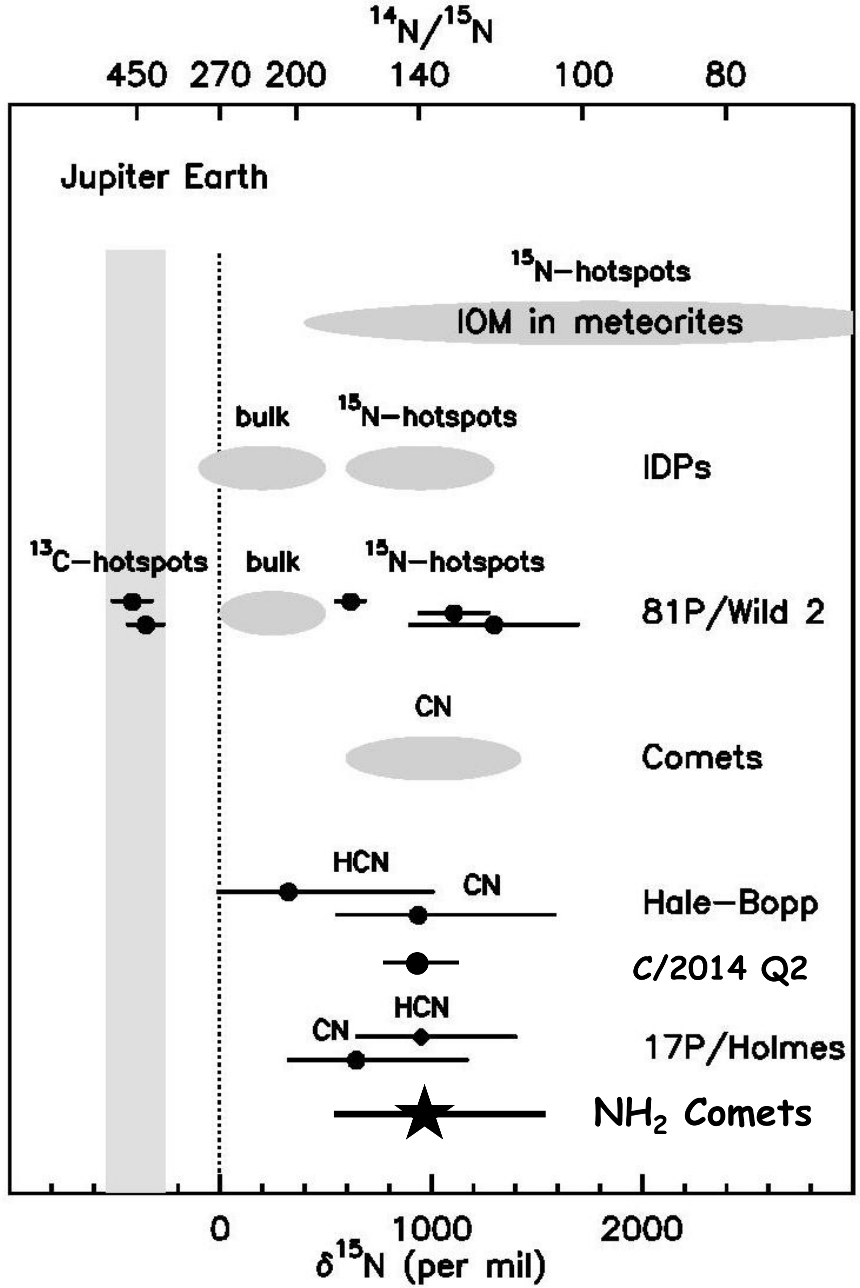


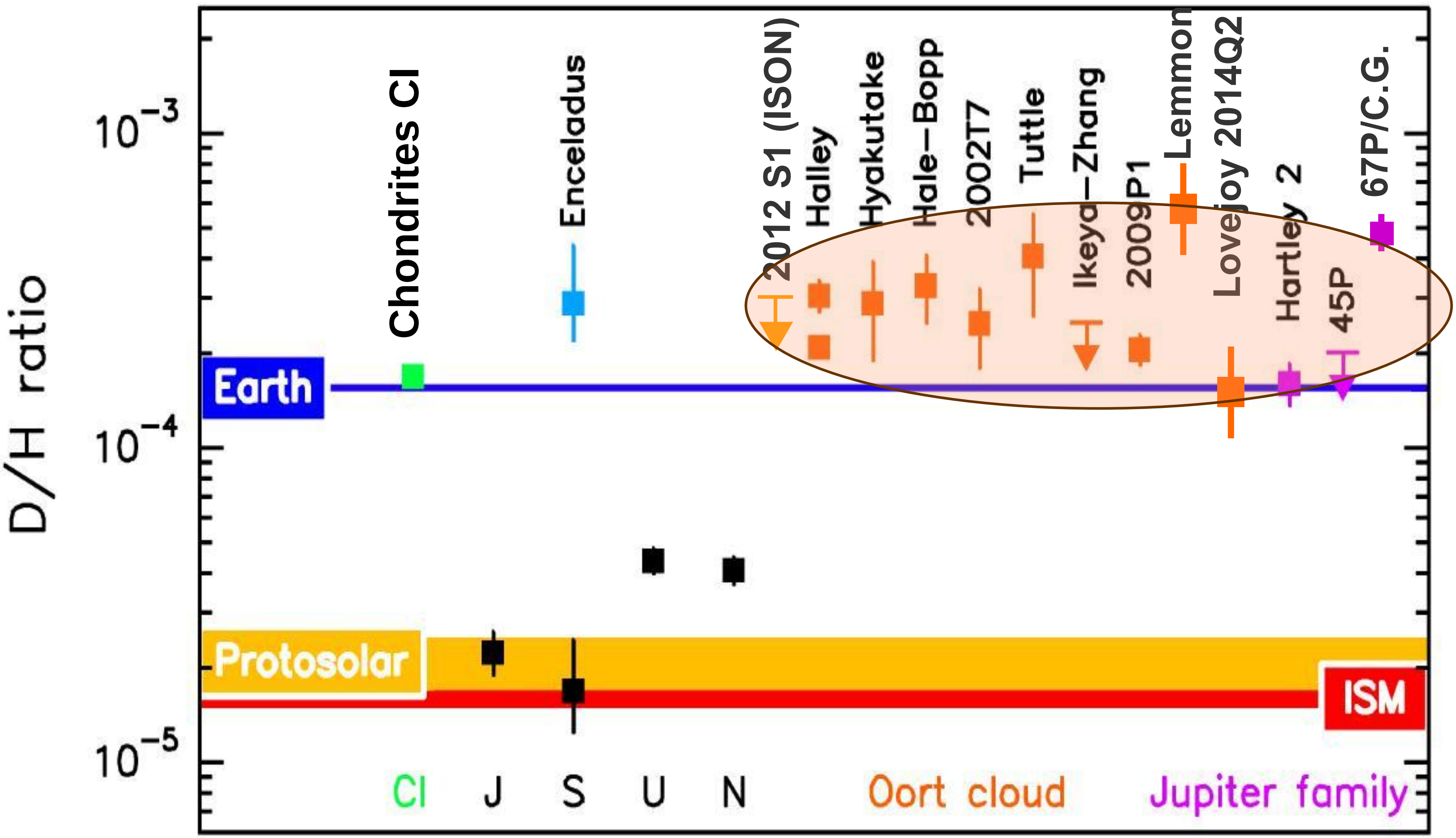
Figure 1. Distribution of the D/H ratio in the carbonaceous meteorites ($N = 106$).

D/H: (1) **up to 3xEarth** (carbon phase, not from water of 81P)
(2) **1 to 50xEarth** in CP-IDPs (likely related to comets) organic matter
(3) enrichment of up to **10-30xEarth** in D-rich areas

$^{14}N/^{15}N$: (1) = **280-180** (bulk, up to **120±20** in some small « hot spots »)
(2) = **236** average, 118 to 300 (^{15}N rich to poor spots)
(3) ^{15}N-rich and ^{15}N-poor regions (Duprat et al. 2014)

$^{14}N/^{15}N$ in the solar system, comets





Hartogh et al. (2011) Nature; Bockelée-Morvan et al. (2012), A&A; Lis et al. (2013), ApJL (HERSCHEL), Altwegg et al. (2015) Science 347 (ROSETTA); Biver et al. (2016) A&A (IRAM); Gibbs et al. (2016) ApJ (IRTF)