New insights on Saturn's formation from its nitrogen isotopic composition

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ABSTRACT

The recent derivation of a lower limit for the ${}^{14}N/{}^{15}N$ ratio in Saturn's ammonia, which is found to be consistent with the Jovian value, prompted us to revise models of Saturn's formation using as constraints the supersolar abundances of heavy elements measured in its atmosphere. Here we find that it is possible to account for both Saturn's chemical and isotopic compositions if one assumes its formation at ~45 K in the protosolar nebula, provided that the O abundance was ~2.6 times protosolar in its feeding zone. To do so, we used a statistical thermodynamic model to investigate the composition of the clathrate phase that formed during the cooling of the protosolar nebula and from which the building blocks of Saturn were agglomerated. We find that Saturn's O/H is at least ~34.9 times protosolar and that the corresponding mass of heavy elements (~43.1 M_{\oplus}) is within the range predicted by semi-convective interior models.

Subject headings: planets and satellites: individual (Saturn) – planets and satellites: formation – planets and satellites: composition – planet and satellites: atmospheres – protoplanetary disks

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1. Introduction

The measurements of 14 N/ 15 N ratios throughout the solar system can be divided into three categories (see Mandt et al. 2014 and references therein): the solar wind and Jupiter have the lightest ratios, presumed to be representative of the protosolar ratio. Chondrites, grains coming from comet 81P/Wild 2, Earth's mantle and atmosphere, Venus and Mars' mantle have moderately heavy ratios. Saturn's moon Titan, Mars' atmosphere, as well as NH₃ and HCN in comets share the heaviest values.

The recent derivation of a 1-sigma lower limit for the ${}^{14}N/{}^{15}N$ ratio in Saturn's ammonia, which is found to be ~500 from TEXES/IRTF ground-based mid-infrared spectroscopic observations (Fletcher et al. 2014), prompts us to revise models of Saturn's formation that previously only used the supersolar abundances of heavy elements measured in the atmosphere as constraints. This lower limit is formally consistent with the ${}^{14}N/{}^{15}N$ ratio (~435) measured by the Galileo probe at Jupiter (Fletcher et al. 2014) and implies that the two giant planets were essentially formed from the same nitrogen reservoir in the nebula, which is N₂ (Owen et al. 2001; Fletcher et al. 2014). Any scenario depicting Saturn's formation should match the ${}^{14}N/{}^{15}N$ ratio measured in its atmosphere and be consistent with disk's temperatures greater than 30 K in the giant planets formation region. Lower temperatures have only been observed in regions located beyond ~30 AU in circumstellar disks (Qi et al. 2013).

Two scenarios of Saturn's formation, aiming at matching the supersolar volatile abundances measured in its envelope, have been proposed. Both approaches determine the composition of the planet's building blocks from a simple clathrate formation model and assume that all elements were in protosolar abundances in the disk's gas phase. The first scenario, proposed by Hersant et al. (2008), assumes that Saturn formed at ~40–50K in the protosolar nebula (hereafter PSN). In their model, NH₃ was trapped in planetesimals, while the dominant N molecule in the PSN, N₂, remained well mixed with H₂ until the gas collapsed onto the core of the planet. This scenario is now ruled out because it suggests that Saturn's supersolar N abundance essentially results from the delivery of NH₃ trapped in solids, implying that its ¹⁴N/¹⁵N ratio should be substantially lower than the Jovian value (Hersant et al. 2008).

Alternatively, Mousis et al. (2009b) proposed that Saturn formed at a cooler temperature in the disk. In this scenario, planetesimals were agglomerated from a mixture of clathrates and pure ices condensed close to ~20 K, implying that both NH₃ and N₂ were trapped in solids. Their model is consistent with the measured $^{14}N/^{15}N$ ratio since N₂ remains the main nitrogen reservoir delivered to Saturn. However, the formation of Saturn at such a low temperature in the PSN is questionable as the heating of the disk by proto-Sun's UV radiation might prevent the temperature from decreasing down to ~ 40 K at 10 AU (D'alessio et al. 1998).

Here we find that it is possible to account for both Saturn's chemical and isotopic compositions if one assumes its formation at ~45 K in the PSN, provided that the O abundance was ~2.6 times protosolar in its feeding zone. To do so, we used a statistical thermodynamic model (Mousis et al. 2010, 2012) to investigate the composition of the clathrate phase that formed during the cooling of the PSN from the most abundant gaseous volatiles. These clathrates agglomerated with the other condensates and formed the building blocks of Saturn. A fraction of these planetesimals accreted in the growing Saturn dissolved in its envelope and subsequently engendered the observed volatile enrichments.

2. Useful elemental abundances measured in Saturn

Table 1 summarizes the abundances of C, N, P, S and O, normalized to their protosolar abundances, and measured in the forms of CH_4 , NH_3 , PH_3 , H_2S (indirect determination) and H_2O in Saturn's atmosphere. Note that the protosolar abundances correspond to the present day solar values corrected from elemental settling in the Sun over the past 4.56 Gyr (Lodders et al. 2009). The abundance of CH_4 has been determined from the analysis of high spectral resolution observations from Cassini/CIRS (Fletcher et al. 2009a). As methane does not condense at Saturn's atmospheric temperatures, its atmospheric abundance can be considered as representative of the bulk interior. The NH₃ abundance is taken from the range of values derived at the equator by Fletcher et al. (2011) from Cassini/VIMS 4.6–5.1 μm thermal emission spectroscopy. The measured NH_3 abundance may be considered as a lower limit since the condensation level of NH₃-bearing volatiles may be deeper than the sampled regions (Atreya et al. 1999, 2014), implying that there could be a large reservoir of ammonia hidden below the condensate cloud decks. PH_3 has been determined remotely in Saturn from Cassini/CIRS observations at 10 μ m (Fletcher et al. 2009b). PH₃ is a disequilibrium species and its bulk abundance may be higher than the inferred value, depending on the rates of vertical mixing and photochemical destruction. The H₂S abundance is quoted from the indirect determination of Briggs & Sackett (1989) from radio observations but remains highly uncertain.

Species	$Measurements^{(\star)}$	(1)	(2)
0	_	34.9	14.2
\mathbf{C}	$9.6 \pm 1.0^{(a)}$	8.6	8.6
Ν	$2.8 \pm 1.1^{(b)}$	7.5	5.9
\mathbf{S}	$12.05^{(c)}$	5.2	7.1
Р	$11.2 \pm 1.3^{(d)}$	10.4	14.2
Ar	_	1.9	5.5
Kr	—	8.3	7.5
Xe	—	10.4	14.2

Table 1: Observed and calculated enrichments in volatiles in Saturn

Notes. (1) and (2) correspond to the full and limited clathration scenarios, respectively. Saturn's formation temperature is considered at ~45 K (full clathration scenario) and 22 K (limited clathration scenario). The observed values are derived from ^(a)Fletcher et al. (2009a), ^(b)Fletcher et al. (2011), ^(c)Briggs & Sackett (1989) and ^(d)Fletcher et al. (2009b), using the protosolar abundances of Lodders et al. (2009). ^(*)Error is defined as $(\Delta E/E)^2 = (\Delta X_{Saturn}/X_{Saturn})^2 + (\Delta X_{\odot}/X_{\odot})^2$.

3. Model description

In our model, the volatile phase incorporated in planetesimals is composed of a mixture of pure ices, stoichiometric hydrates (such as NH_3-H_2O hydrate) and multiple guest clathrates that crystallized in the form of microscopic grains at various temperatures in the outer part of the disk. Our model is based on the assumption that planetesimals have grown from the agglomeration of these icy grains due to collisional coagulation (Weidenschilling 1997). Here, the clathration process stops when no more crystalline water ice is available to trap the volatile species and then only pure condensates can form subsequently if the disk cools down to very low temperatures. The process of volatile trapping in icy grains is calculated using the equilibrium curves of hydrates and pure condensates, our model determining the equilibrium curves and compositions of MG clathrates, and the thermodynamic path detailing the evolution of temperature and pressure at the current location of Saturn in the protosolar nebula.

Our computations are based on a predefined initial gas phase composition in which all elemental abundances, except that of oxygen in some circumstances (see Sec. 5), are protosolar (Lodders et al. 2009). We assume that O, C, and N exist only under the form of H_2O , CO, CO_2 , CH_3OH , CH_4 , N_2 , and NH_3 . Hence, once the gaseous abundances of elements are defined, the abundances of CO, CO_2 , CH_3OH , CH_4 , N_2 , and NH_3 are determined from the adopted CO:CO₂:CH₃OH:CH₄, and N₂:NH₃ gas phase molecular ratios. Finally, once the abundances of these molecules are fixed, the remaining O gives the abundance of H₂O. We set CO:CO₂:CH₃OH:CH₄ = 10:4:1.67:1 in the gas phase of the disk, values that are consistent with interstellar medium (ISM) measurements considering the contributions of both gas and solid phases in the lines of sight (Pontoppidan et al. 2006; Öberg et al. 2011) and measurements of production rates of molecules in Comet C/1995 O1 Hale-Bopp (Bockélee-Morvan et al. 2004). In addition, S is assumed to exist in the form of H₂S, with H₂S:H₂ = $0.5 \times (S:H_2)_{\odot}$, and other refractory sulfide components (Pasek et al. 2005). We finally consider N₂:NH₃ = 10:1 in the nebula gas-phase, a value predicted by thermochemical models of the solar nebula (Lewis & Prinn 1980).

The process of volatile trapping in planetesimals formed in the feeding zone of Saturn follows the approach depicted in Mousis et al. (2012) who used a statistical thermodynamic model to compute the composition of multiple guest clathrates formed in the PSN. We refer the reader to this paper for further information on the model. Here, our computations have been made in the case of formation of Structure I multiple guest clathrates in the PSN because CO, CO_2 and H_2S , which are the most abundant volatiles in the gas phase, also individually form Structure I clathrates.

Figure 1 shows two cases for the compositions of planetesimals condensed in Saturn's feeding zone and represented as a function of their formation temperature. In both cases, NH₃ forms NH₃-H₂O hydrate and CH₃OH is assumed to condense as pure ice in the PSN because of the lack of thermodynamic data concerning its associated clathrate. In the first case (full clathration), all volatiles (except NH₃ and CH₃OH) are trapped in the clathrate phase as a result of an initial supersolar oxygen abundance ($\sim 2.6 \times (O/H)_{\odot}$) in Saturn's feeding zone. In the second case (limited clathration), we used a protosolar abundance for oxygen, implying that the budget of available crystalline water is not sufficient to trap all volatiles in clathrates. In this case, significant fractions of CO, N₂ and Ar form pure ices if the disk cools down to very low temperatures (~ 20 K), instead of being trapped in clathrates, as it is the case for full volatile clathration. For example, Ar, N₂ and CO become substantially trapped in the clathrate phase at ~ 38 , 45, and 48 K in the PSN, respectively. In contrast, these species form pure ices in the 22–26 K range in the PSN.

Assuming that the composition of the icy phase of planetesimals computed with our model is representative of that of Saturn's building blocks, the precise adjustment of their mass accreted by the forming Saturn and vaporized into its envelope allows us to reproduce the observed volatile enrichments. Here, because of the lack of reliable measurements, our fitting strategy is to match the minimum carbon enrichment measured in Saturn. By doing so, this allows us to maintain the mass of solids accreted into Saturn's envelope as small as

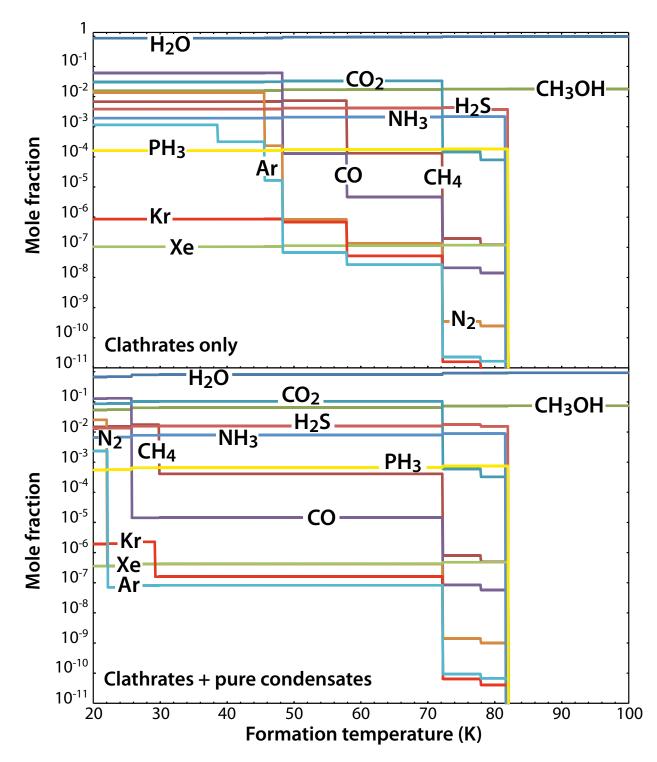


Fig. 1.— Composition of the volatile phase incorporated in planetesimals formed beyond the snow line in the PSN as a function of their formation temperature (full clathration case). Top: volatiles are fully trapped in clathrates in the PSN. Bottom: ices are composed of a mixture of pure condensates and clathrates (limited clathration case).

possible in order to be compared to the mass of heavy elements predicted by interior models.

4. Results

Figure 2 shows the evolution of the N₂/NH₃ ratio in Saturn as a function of its formation temperature, and assuming that it was equal to 10 in the PSN prior to planetesimals formation. Depending on the temperature considered for Saturn's formation, contributions of both N₂ and NH₃ in solid and gaseous phases have been taken into account in our computation. When not trapped or condensed, the species collapse with the nebula gas onto the forming planet and form its gaseous envelope. NH₃ is always in solid form at T < 80 K. In the full clathration case, the maximum temperature of Saturn's formation yielding N₂ \gg NH₃ in the envelope is ~45 K. Above this temperature, N₂ remains essentially in gaseous form. In contrast, in the limited clathration case, N₂ dominates in Saturn only at formation temperatures lower than ~22 K, a value corresponding to the condensation temperature of this species in the PSN. A comparison between the two cases shows that the full volatile clathration favors a higher N₂/NH₃ in Saturn at temperatures below ~45 K. In both situations, as illustrated by Fig. 2, the amount of N₂ supplied to Saturn in gaseous form is less than that of NH₃ in solid form.

Figure 3 represents the volatile enrichments in Saturn calculated from the fit of the minimum C abundance observed in the atmosphere (~8.6 times protosolar – see Table 1), and assuming the planetesimals compositions displayed by Fig. 1. In the full clathration scenario and in the PSN temperature range ($T \leq 45$ K) consistent with the 14 N/ 15 N constraint, we find that N is 7.5 times more enriched than the protosolar value in Saturn's atmosphere, a value higher than the maximum inferred one (3.9 times protosolar), but still lower than the measured C enrichment (9.6 ± 1 times protosolar). In this case, O is predicted to be at least ~34.9 times more enriched than the protosolar value in Saturn's envelope. Interestingly, the calculated P enrichment (~10.4 times protosolar) matches the measured value (11.2 ± 1.3 times protosolar). S is found 5.2 times more enriched than the protosolar).

In the limited clathration scenario and in the PSN temperature range ($T \leq 22$ K) consistent with the 14 N/ 15 N constraint, N and S are found to be 5.9 and 7.1 times more enriched than their protosolar values, respectively. Both calculated enrichments are closer to the observed enrichments than those derived in the full clathration scenario. On the other hand, P is more enriched (14.2 times protosolar) than the observed value (11.2 ± 1.3 times protosolar). In this case, O/H is also found to be at least 14.2 times more enriched in Saturn than the protosolar value. Regardless the variations of the fits accuracy between the two

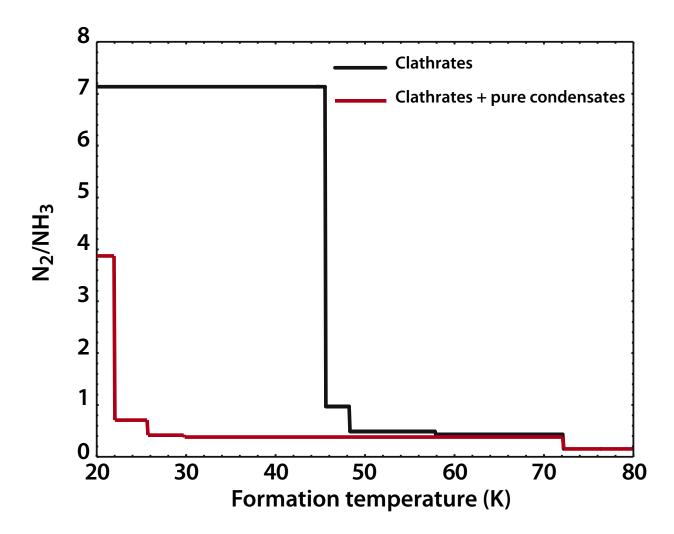


Fig. 2.— N_2/NH_3 ratio in the envelope of Saturn as a function of its formation temperature in the cases of building blocks agglomerated from clathrates (black curve) and from a mixture of clathrates and pure condensates (red curve).

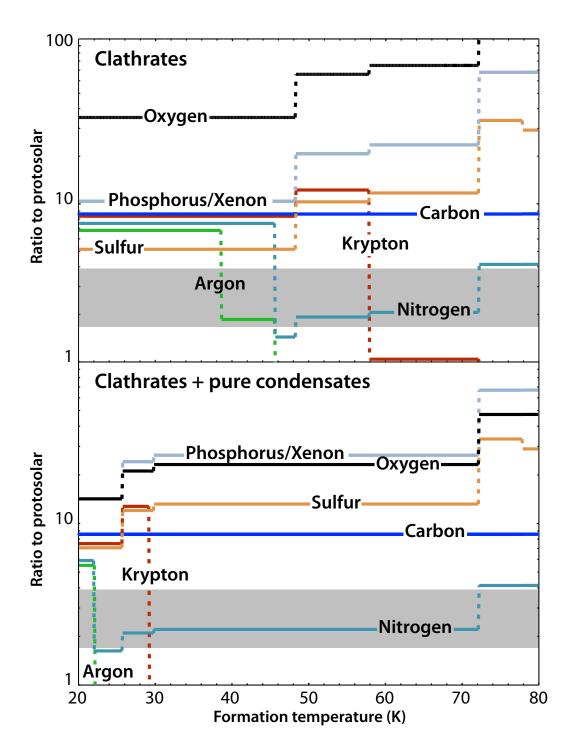


Fig. 3.— Volatile enrichments computed in Saturn's atmosphere as a function of its formation temperature. The results have been fitted to the minimum value of carbon enrichment measured in Saturn's atmosphere (see Table 1). Top panel: volatiles fully frapped in clathrates in Saturn's feeding zone (full clathration scenario). Bottom panel: volatiles are delivered to Saturn in the form of a mixture of pure condensates and clathrates (limited clathration scenario). In both cases, P and Xe enrichments appear superimposed and the grey area represents the uncertainties on the N measurement.

scenarios, it must be borne in mind that only the C determination is fairly robust and that it is not guaranteed that the N, P and S measurements are representative of Saturn's bulk composition. Hence, it is difficult to derive firm conclusions on which of the two scenarios – full or limited clathration – is the most likely, based on the fits of the existing measurements solely. Table 1 summarizes the enrichments calculated in the two different scenarios and gives predictions for the volatile species that have not been yet detected (Ar, Kr, Xe) or those whose sampling still needs to be investigated (O, N, S, P).

Figure 4 shows that ~27.9 and 15.2 M_{\oplus} of ices (including ~21.9 and 8.0 M_{\oplus} of water) are needed in Saturn to match the minimum C enrichment in the full and limited clathration scenarios at ~45 and 22 K in the PSN, respectively. Our calculated mass must be seen as a minimum because planetesimals may harbor a significant fraction of refractory phase. Assuming an ice-to-rock ratio of ~1 for a protosolar composition gas (Johnson et al. 2012), this implies that ~43.1 and 30.4 M_{\oplus} of heavy elements have been delivered to Saturn in the full and limited clathration scenarios. The mass of heavy elements needed by the full clathration scenario then exceeds the maximum mass of heavy elements predicted in Saturn by homogeneous interior models ($\leq 30 M_{\oplus}$; Nettelmann et al. 2013) while the one required by the limited clathration scenario remains close to the upper limit. On the other hand, both scenarios are consistent with the mass range (26-50 M_{\oplus}) of heavy elements predicted by semi-convective models (Leconte & Chabrier 2012).

5. Discussion

The lower limit for the ${}^{14}N/{}^{15}N$ ratio found by Fletcher et al. (2014) implies that Saturn's nitrogen was essentially accreted in N₂ form at its formation time. However, this condition is not sufficient to match the measured ${}^{14}N/{}^{15}N$ ratio: our calculations suggest that N₂ must have been accreted in solid form in Saturn, in order to match the observed C enrichment, otherwise NH₃ would still remain the main N-bearing reservoir in the envelope.

We have explored two hypotheses to simultaneously account for the ¹⁴N/¹⁵N measurement and the volatile enrichments in Saturn by varying the O/H ratio in the giant planet's feeding zone. Both possibilities were investigated by using a statistical thermodynamic approach allowing us to compute the composition of clathrates formed in the PSN. In the first case (full clathration scenario), we assumed that oxygen was sufficiently abundant (~2.6 × (O/H)_☉) to trap all volatiles as clathrates in Saturn's feeding zone (except NH₃ which forms a stochiometric hydrate and CH₃OH due to the lack of thermodynamic data concerning its associated clathrate), leading to N₂ trapping in planetesimals at ~45 K in the PSN. In the second case (limited clathration scenario), we assumed that the O abundance was

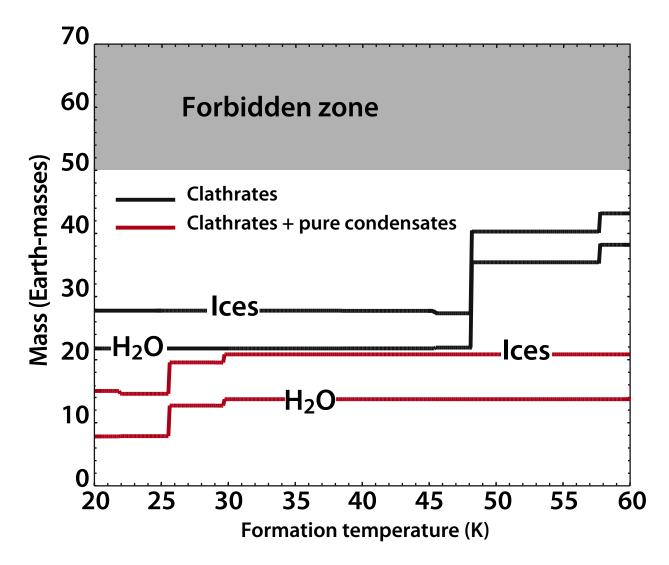


Fig. 4.— Masses of ices and water needed to be accreted in Saturn's envelope in order to match the minimum fit of carbon measurement as a function of the planet's formation temperature. Two different cases are considered: full clathration (black curves) and limited clathration (red curves). In each case, the masses of water and ices (including water) have been computed as a function of temperature. The forbidden zone corresponds to masses of heavy elements above the maximum value predicted in Saturn by semi-convective models (~50 M_{\oplus} ; Leconte & Chabrier 2012).

protosolar, implying that planetesimals were agglomerated from a mixture of clathrates and pure condensates. The PSN had to cool down to ~ 22 K in order to allow the trapping of solid N₂ in planetesimals. Both scenarios match the measured volatile enrichments at the same level of confidence, given the uncertainties concerning their representativity of Saturn's bulk composition, in particular for N and S. The *in situ* measurements of O and Ar may allow disentangling between these two scenarios. O is twice more enriched in Saturn in the full clathration scenario (~ 34.9 times protosolar) than in the limited clathration scenario (~ 14.2 times protosolar). Also, Ar is predicted to be moderately enriched (1.9 times protosolar) at ~ 45 K in the full clathration scenario while it is substantially supersolar (5.5 times protosolar) at ~ 22 K in the limited clathration scenario.

On the other hand, the full clathration hypothesis is the only scenario allowing Saturn's formation at temperatures consistent with our knowledge of the thermal structure of the PSN. The presence of a supersolar oxygen abundance in the giant planet's feeding zone may be explained via its formation in the neighborhood of Jupiter, close to the water iceline location at earlier epochs of the PSN. At this location, the abundance of crystalline water ice may have been enhanced by diffusive redistribution and condensation of water vapor (Stevenson & Lunine 1988; Ali-Dib et al. 2014), thus easing the formation of clathrates. The model of Stevenson & Lunine (1988) predicts enhancements of the surface density of water up to several dozens of times the one derived from a protosolar O abundance.

The full clathration hypothesis matches well the formation scenario of Jupiter proposed by Gautier et al. (2001), where the authors also proposed that the volatiles were fully trapped in clathrates and found O/H ~2.5 × (O/H)_☉ in the giant planet's feeding zone from a simple clathrate formation model. A higher density of water ice in Saturn's feeding zone decreases the ice-to-rock ratio in planetesimals and implies that the icy phase is dominant in the heavy elements accreted by Saturn's envelope. If the full clathration scenario is correct, then it favors the idea that Saturn's interior is heterogeneous and may exhibit a continuous compositional gradient, as illustrated by the model of Leconte & Chabrier (2012). In order to match this model, one needs to argue that a fraction of the heavy elements sedimented onto Saturn's core during its evolution (Fortney & Hubbard 2003, 2004). For example, if all rocks sedimented onto Saturn's core (~15 M_{\oplus}), then the mass of volatiles remaining in the envelope (~28 M_{\oplus}) holds well within the range (10–36 M_{\oplus}) predicted by the semi-convective models of Leconte & Chabrier (2012).

Interestingly, our results are consistent with the fact that NH_3 must be the main primordial reservoir of nitrogen in Titan to explain its current ${}^{14}N/{}^{15}N$ ratio (Mandt et al. 2014). Indeed, formation scenarios predict that Titan's building blocks must have experienced a partial devolatilization during their migration in Saturn's subnebula, which would have induced the loss of the CO, N_2 and Ar captured from the nebula (Mousis et al. 2009a). Hence, Titan's building blocks probably originate from Saturn's feeding zone but they would have been subsequently altered by the subnebula.

Interestingly, notwithstanding the conclusions of the present study, it should be kept in mind that only the *in situ* measurement of O down to the condensation layer of water, and the precise assessment of the C, N, P and the noble gas abundances will be able to shed light on the formation conditions of the ringed planet (Mousis et al. 2014).

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