Geochemical signatures of volatile elements of the Earth's mantle = source or processes?

Insights from F and Cl in arc magmas

Celia Dalou

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Volcanic degassing

Photo diffusée le 29 novembre 2020 par l'agence indonésienne de géologie du mont Lewotolo en éruption, à Lembata, en Indonésie

Volcanic eruption at hte Pu'u Huluhulu volcanic vent on Mount Kilauea, Hawaii.Photograph by james l. Amos, national geographic

Volcanic degassing

- Gas species abundances

	Convergent Plate[b]	Divergent Plate	Hot Spot
T(°C)	768	1130	1140
log fO_2	−14.41	−9.31	−8.82
H_2O	91.9	75.1	75.7
CO_2	4.6	13.1	3.2
H_2	0.5	1.59	0.95
H_2S	0.67	1.01	0.16
SO_2	1.44	7.84	19.4
HCl	0.76	0.42	0.17
HF	0.061	0.42[b]	0.18
CO	0.03	0.6	0.09

[a]Compiled from *Gerlach* [2004].
[b]Based on the proposed ratio HCl/HF = 1 by *Symonds et al.* [1994].

From Hoshyaripour et al. 2012

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Volcanic degassing

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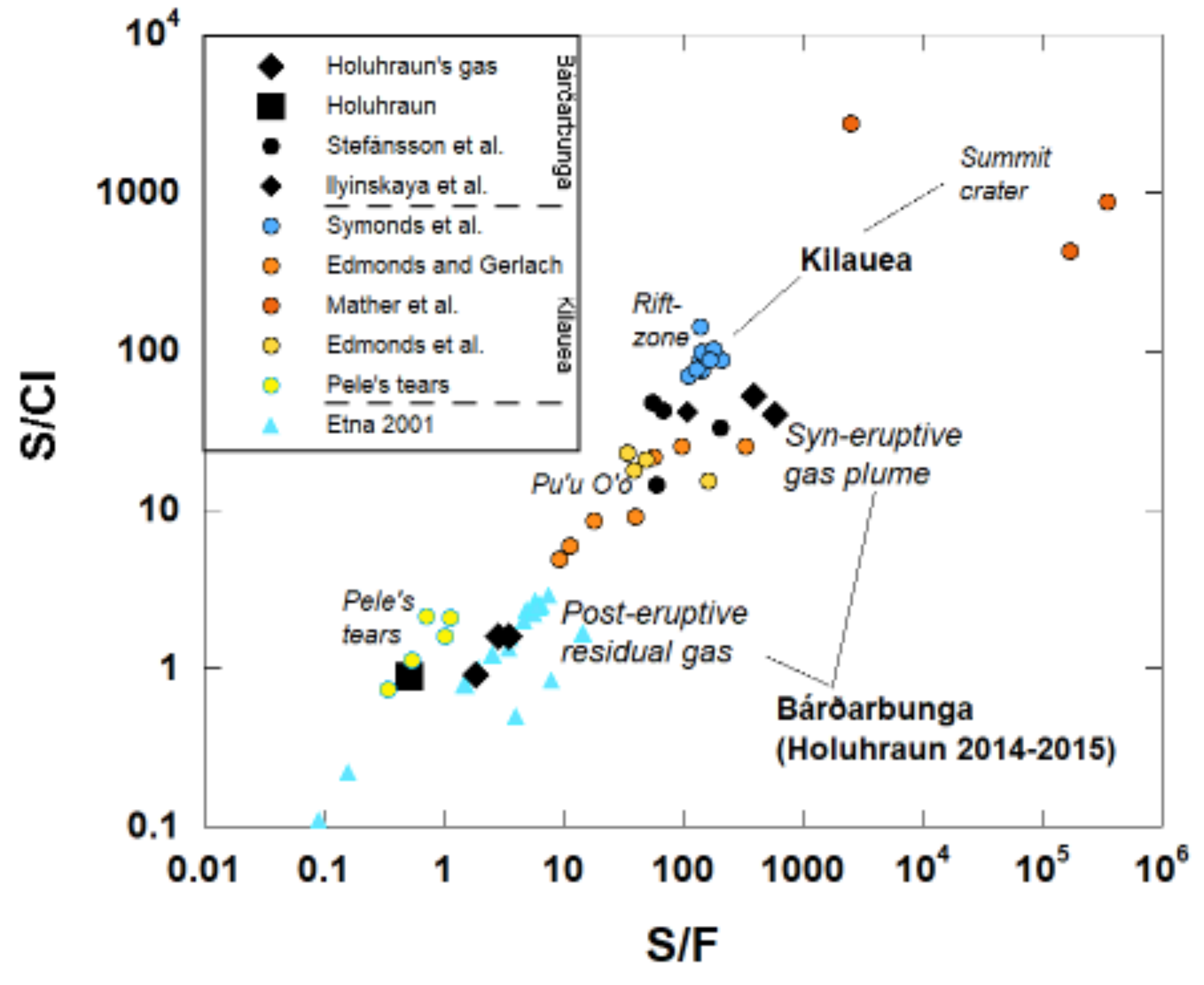

Sigmarsson et al. 2020

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Source or processes ?

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Hydrothermalism

- Gas species abundances

Table 1. Volatile Discharge Through Yellowstone Rivers and Soils, and Abundance in Lavas

	Riverine Discharge[a] ($t\,d^{-1}$)	Diffuse Soil Discharge [b] ($t\,d^{-1}$)	Rhyolitic Melt Inclusions[c] (ppm)
CO_2	546	7,000–33,000	$<$400
S	56	58–275	$<$100
Cl	139	--	1,100
F	18	--	2,000

From Hurwitz and Lowenstern, 2014

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- Gas species abundances

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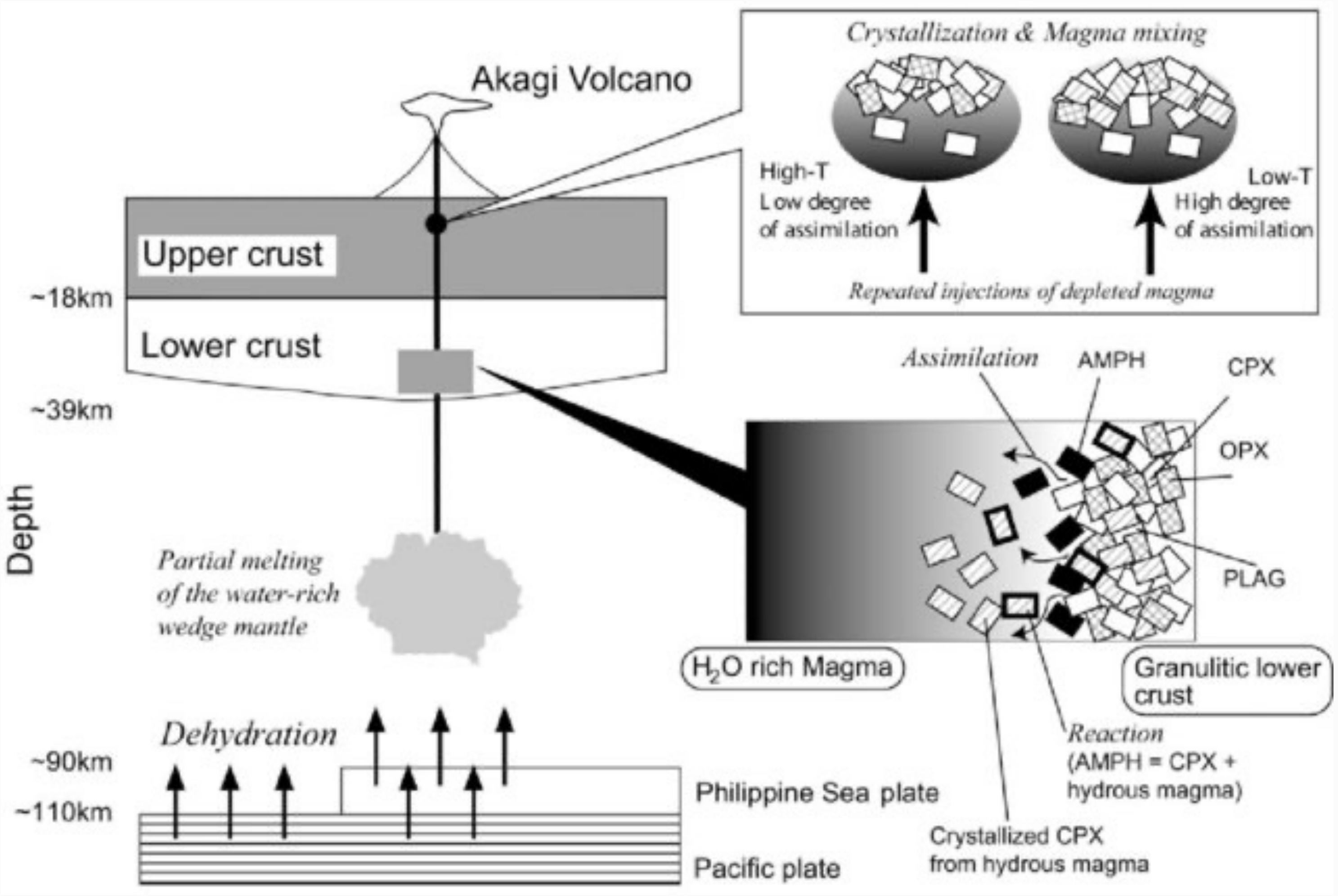
At depth ?

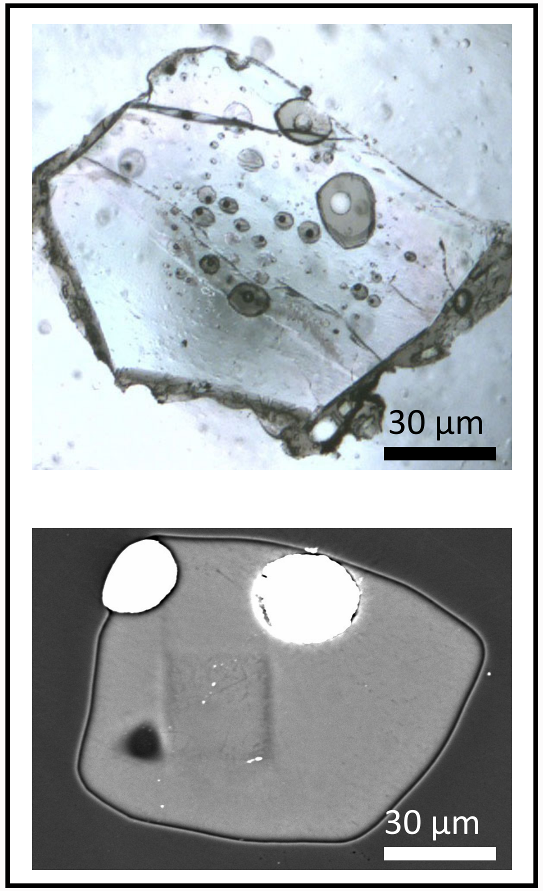
Signature of primary magmas ?



From Kobayashi and Nakamura, 2001

Volatile signatures in arc melt inclusions

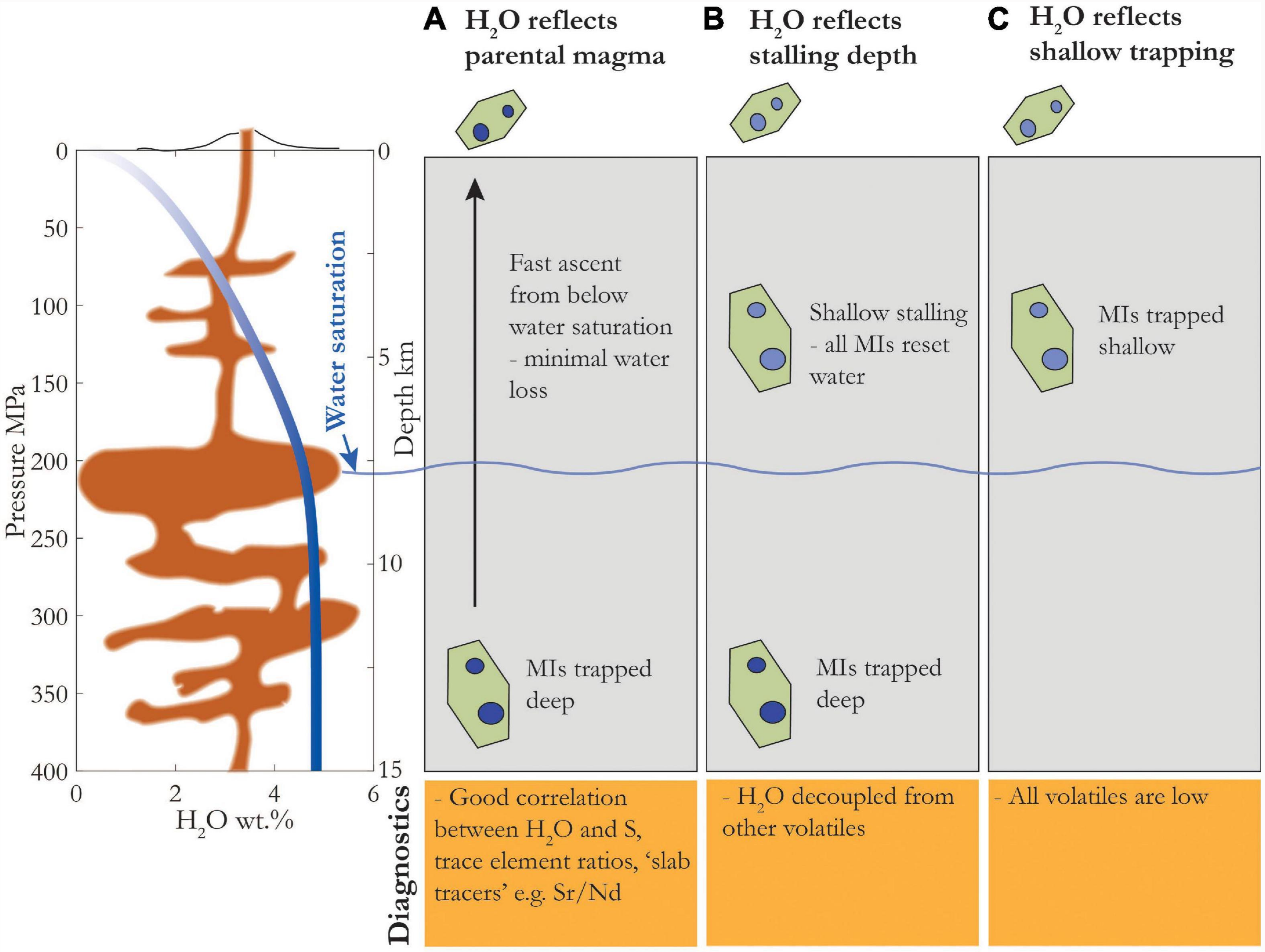
Measured in mineral-hosted melt inclusions from arcs ⇨ Primitive signature of arc magmas



Photos: Marion Le Voyer

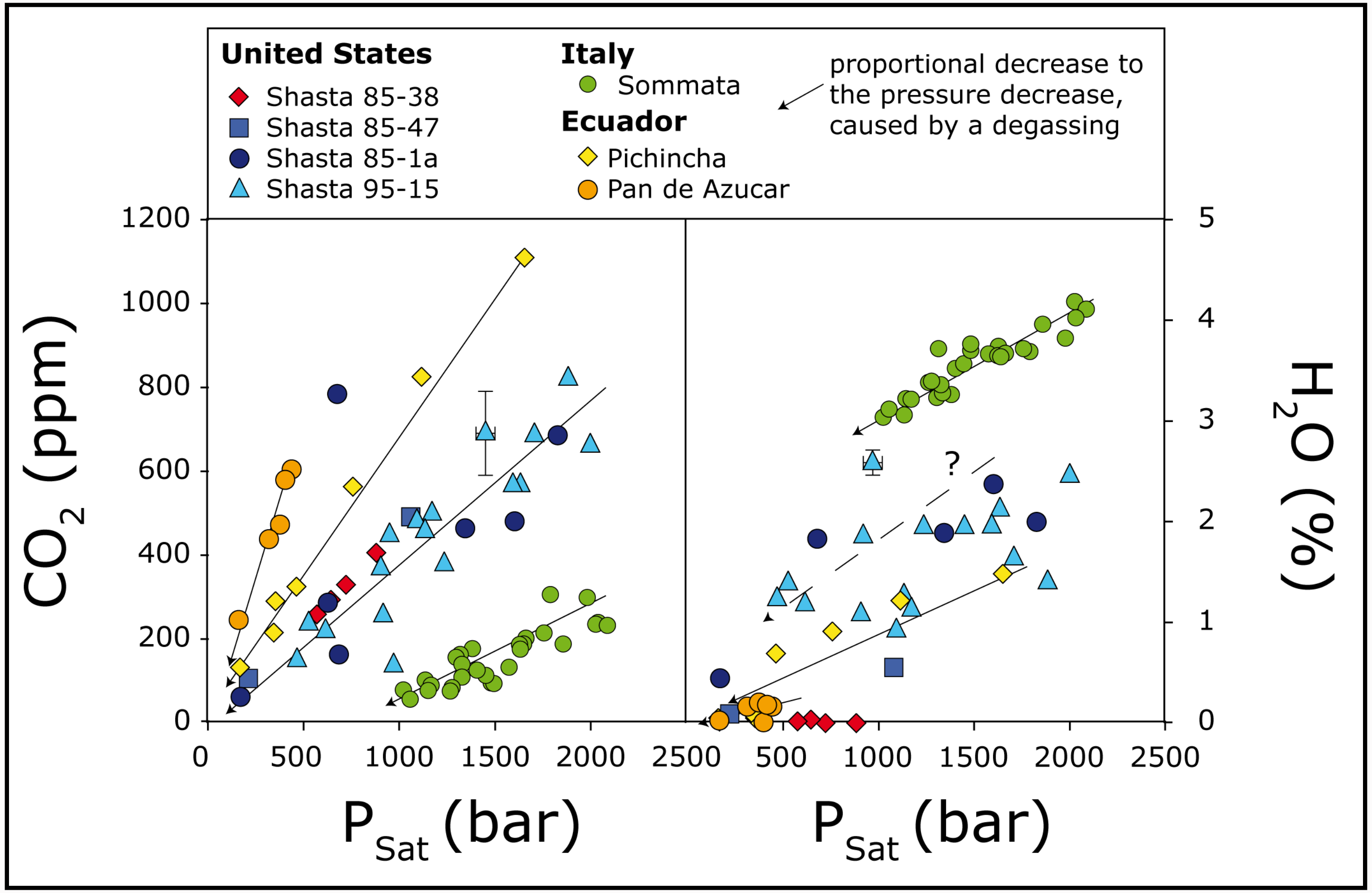
Volatile signatures in arc melt inclusions

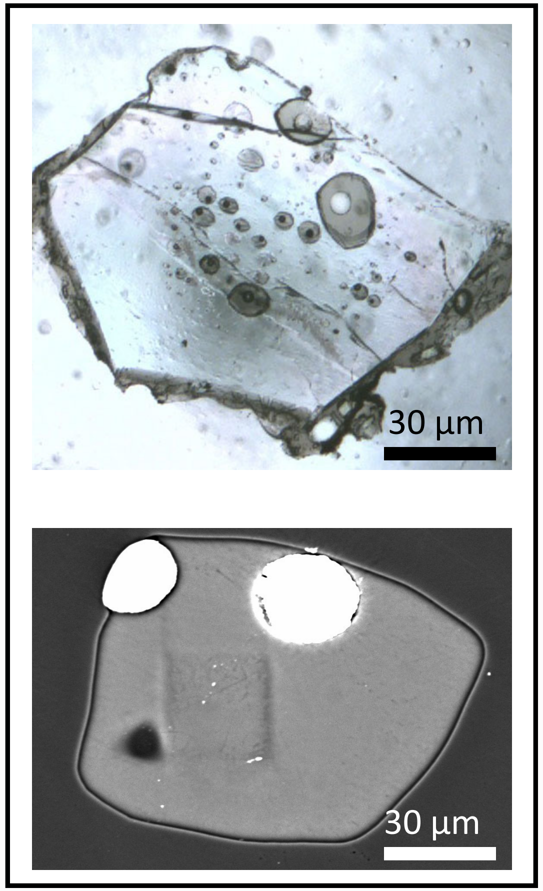
A few precautions:



Barth and Plank, 2001

Volatile signatures in arc melt inclusions





Photos: Marion Le Voyer

Le Voyer, 2009

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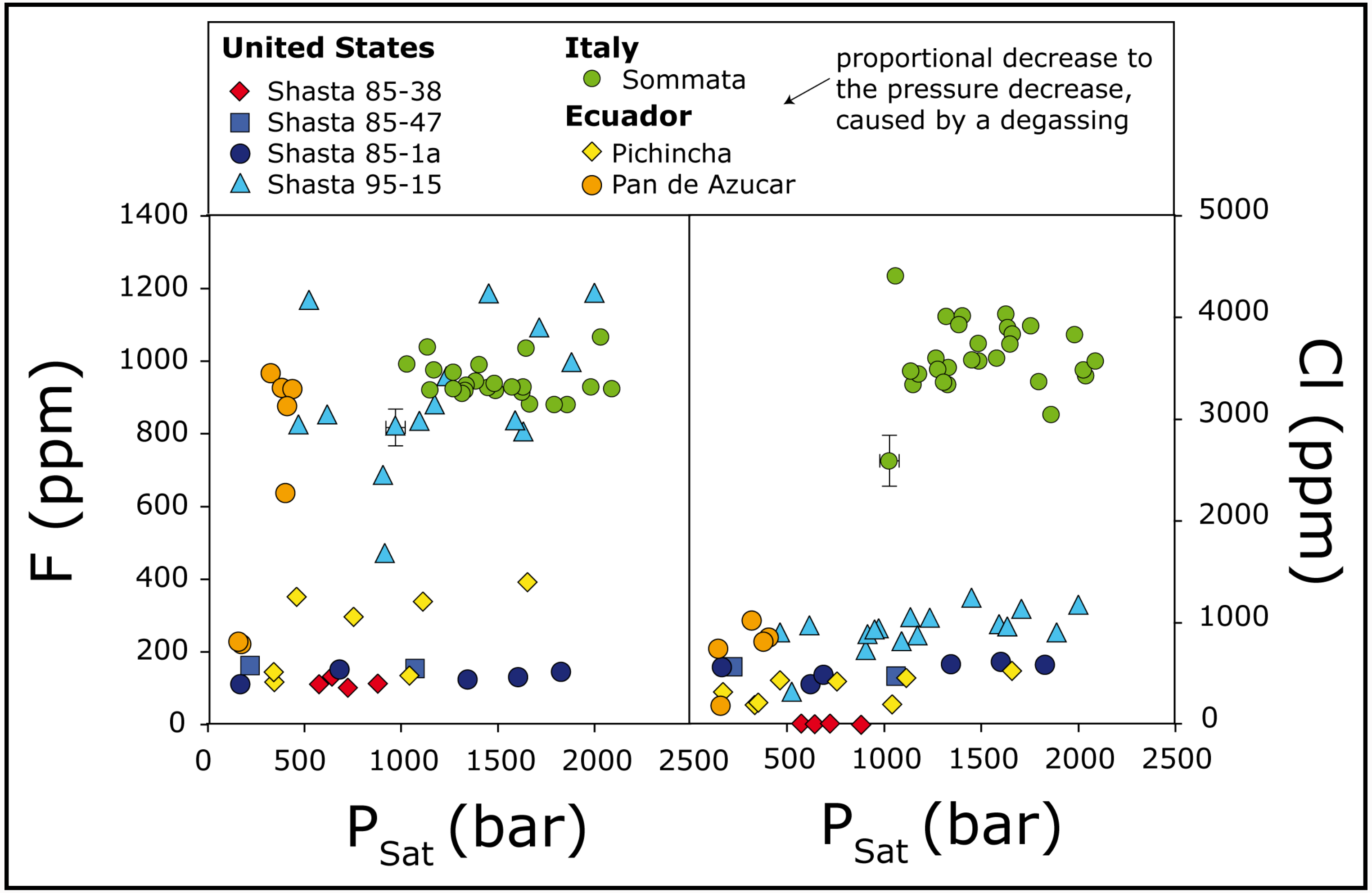
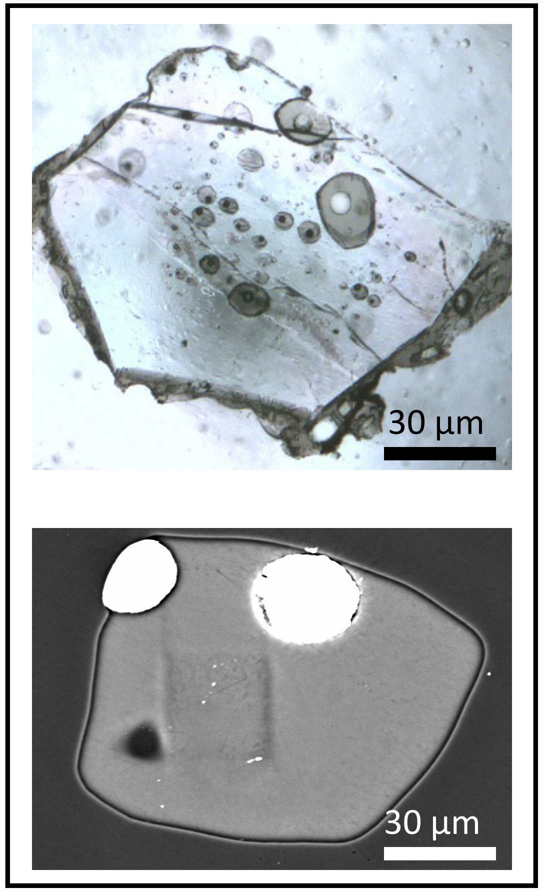




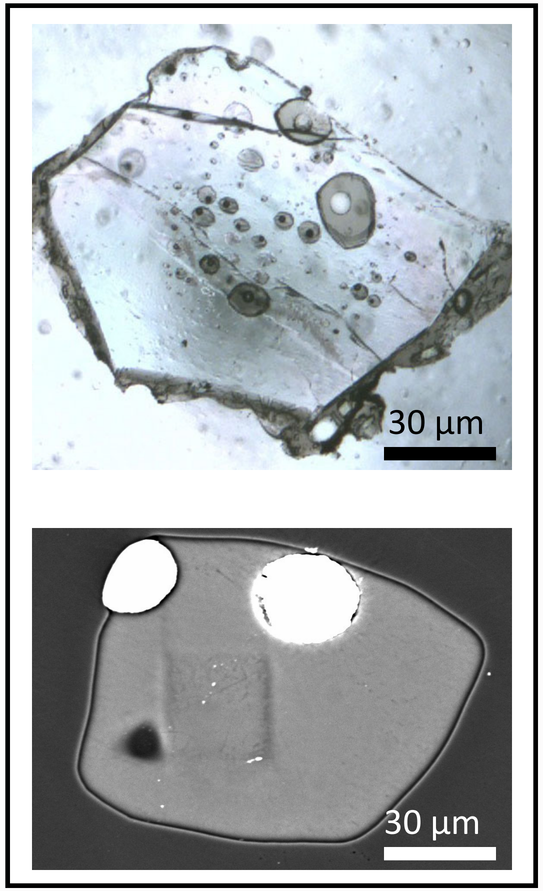
Photos: Marion Le Voyer

Le Voyer, 2009

Volatile signatures in arc melt inclusions

Contrary to H_2O, CO_2 and S,
F and Cl remain undegassed or poorly degassed in melt inclusions.

Their concentration in melt inclusions is most likely **primitive.**



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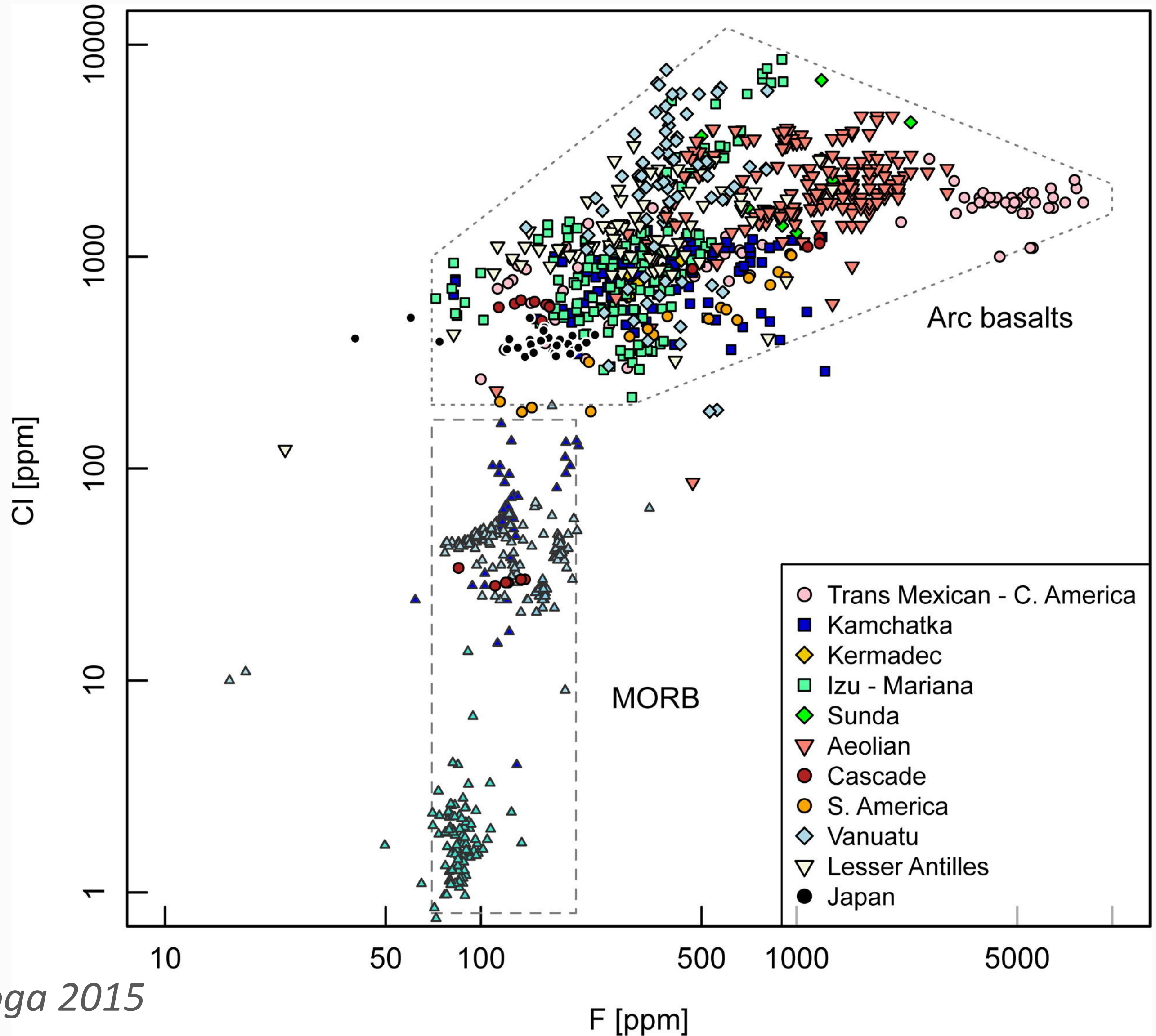
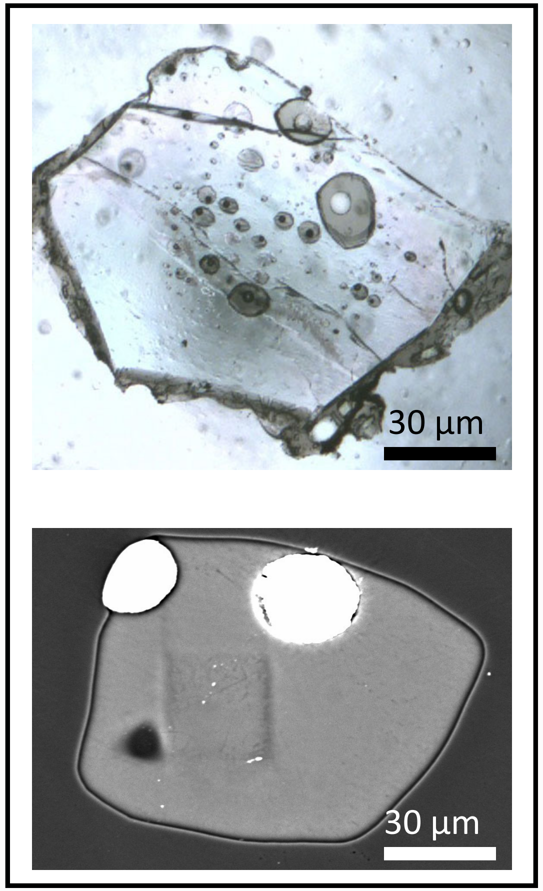
Volatile signatures in arc melt inclusions



Van den Bleeken and Koga 2015



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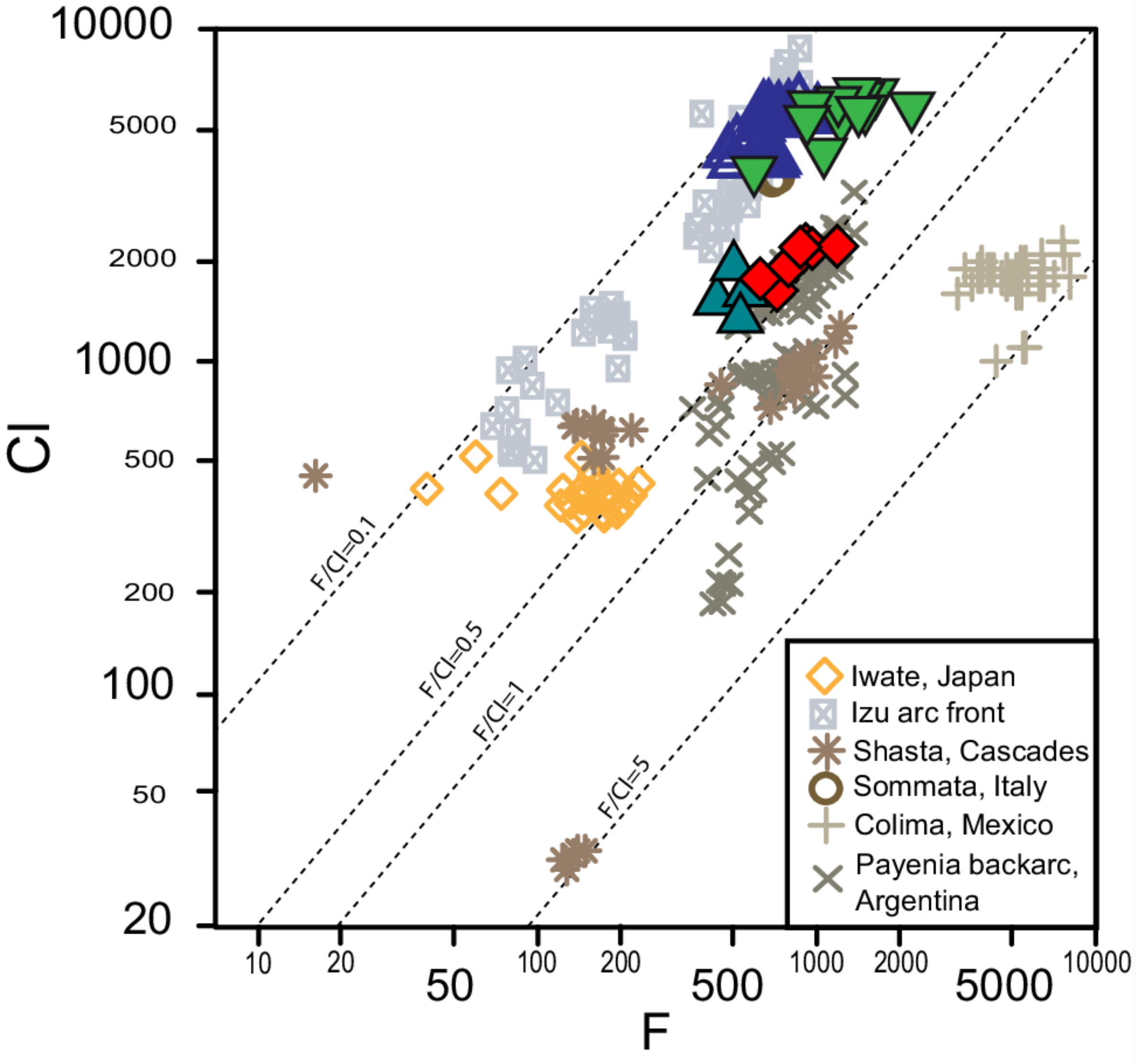
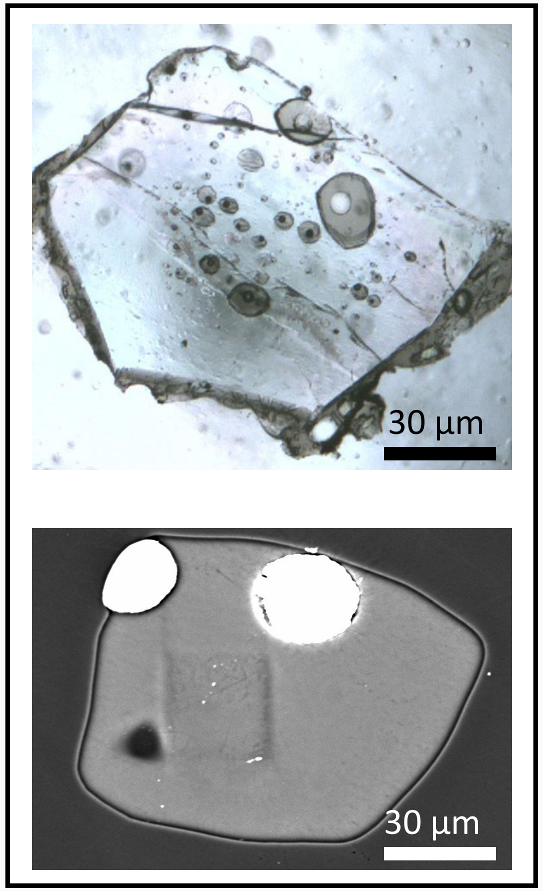


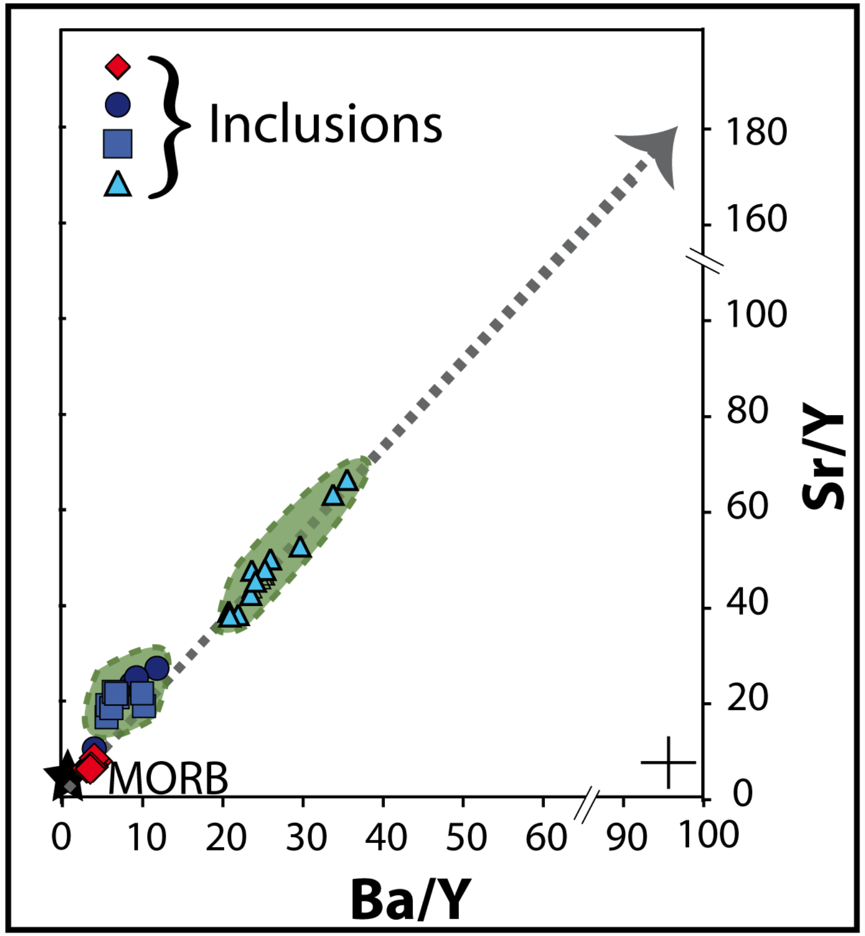
Narvaez et al. 2018

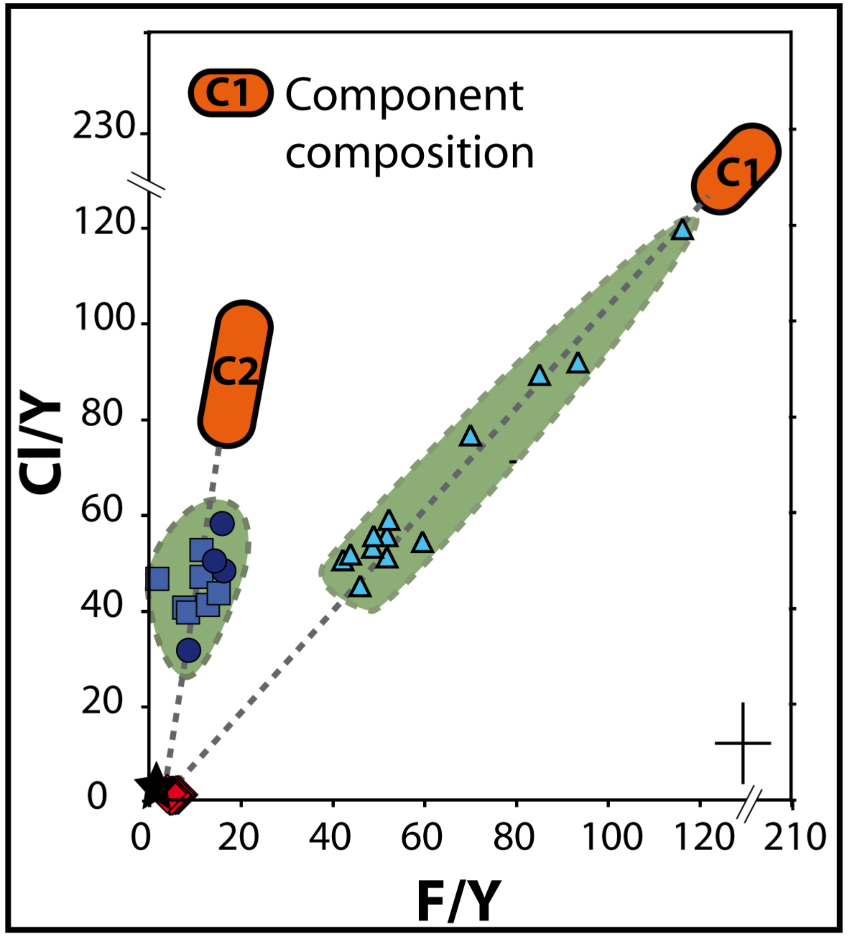


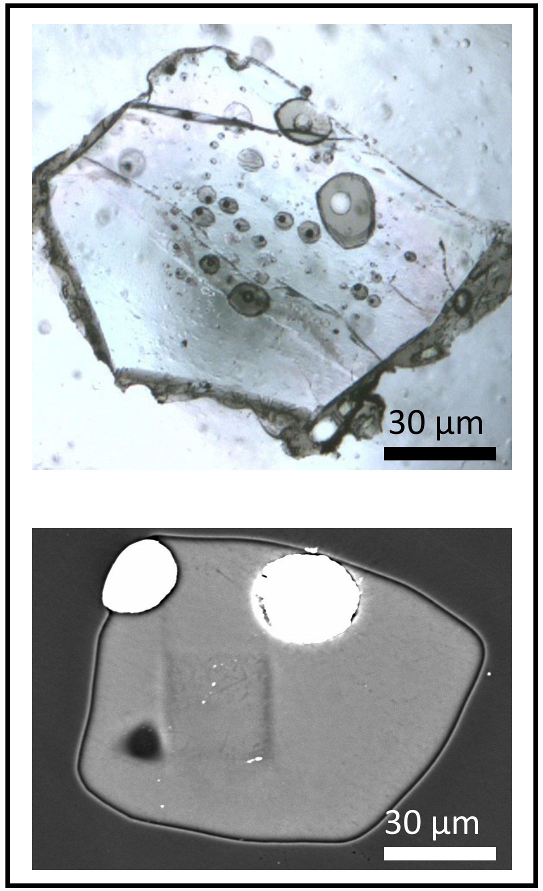
Photos: Marion Le Voyer

Volatile signatures in arc melt inclusions

Within the same volcano (Mount Shasta melt inclusions):



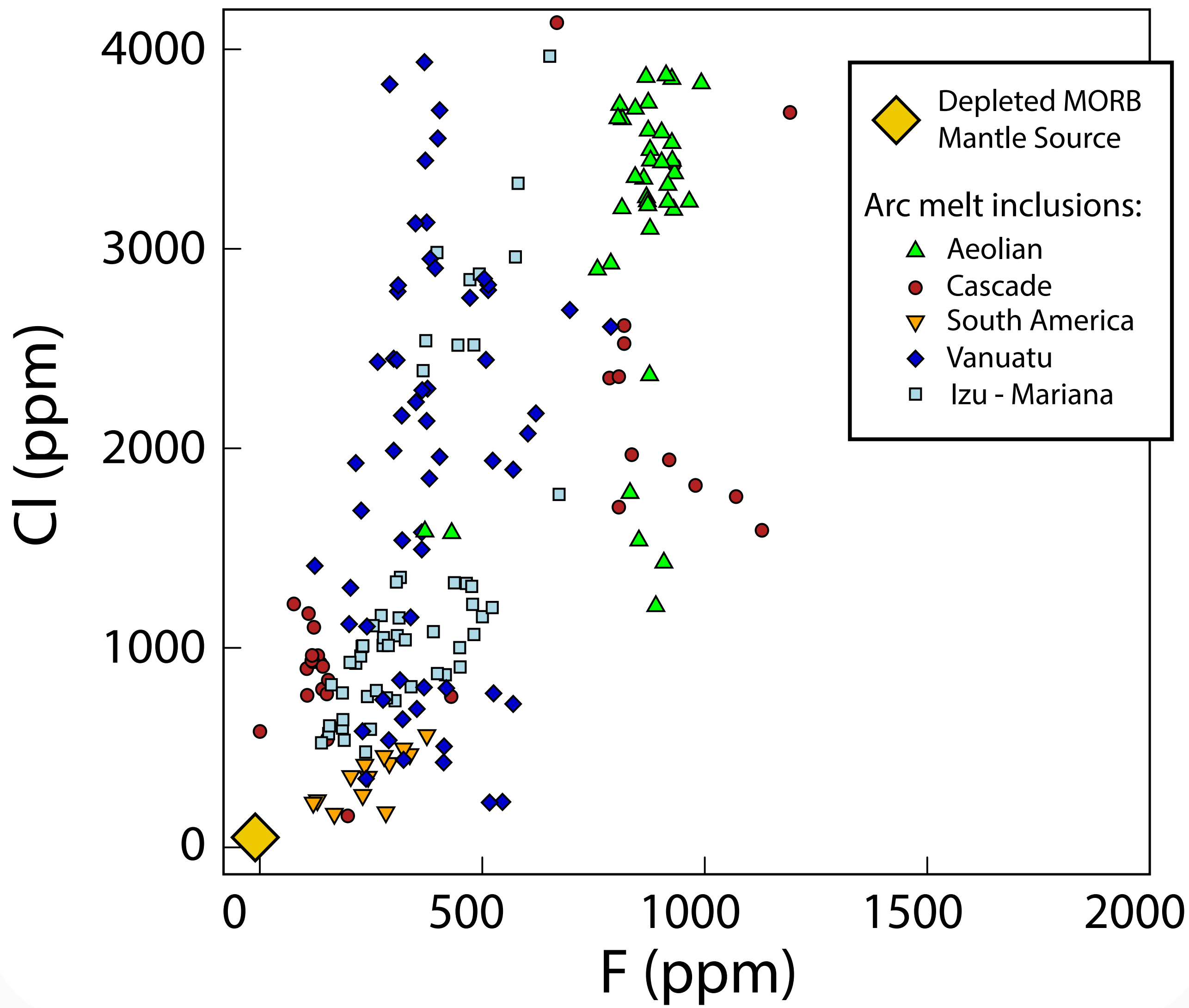


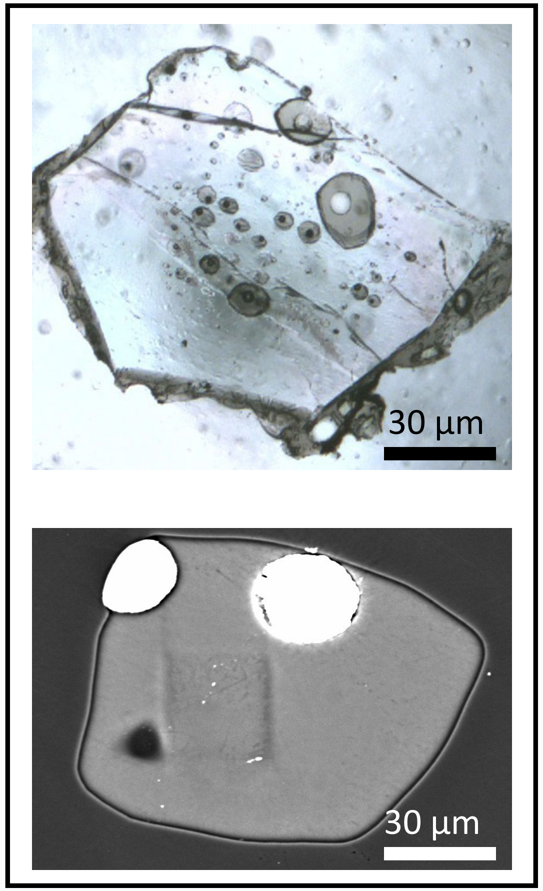


Photos: Marion Le Voyer

Le Voyer et al. 2010

F and Cl in arc magmas





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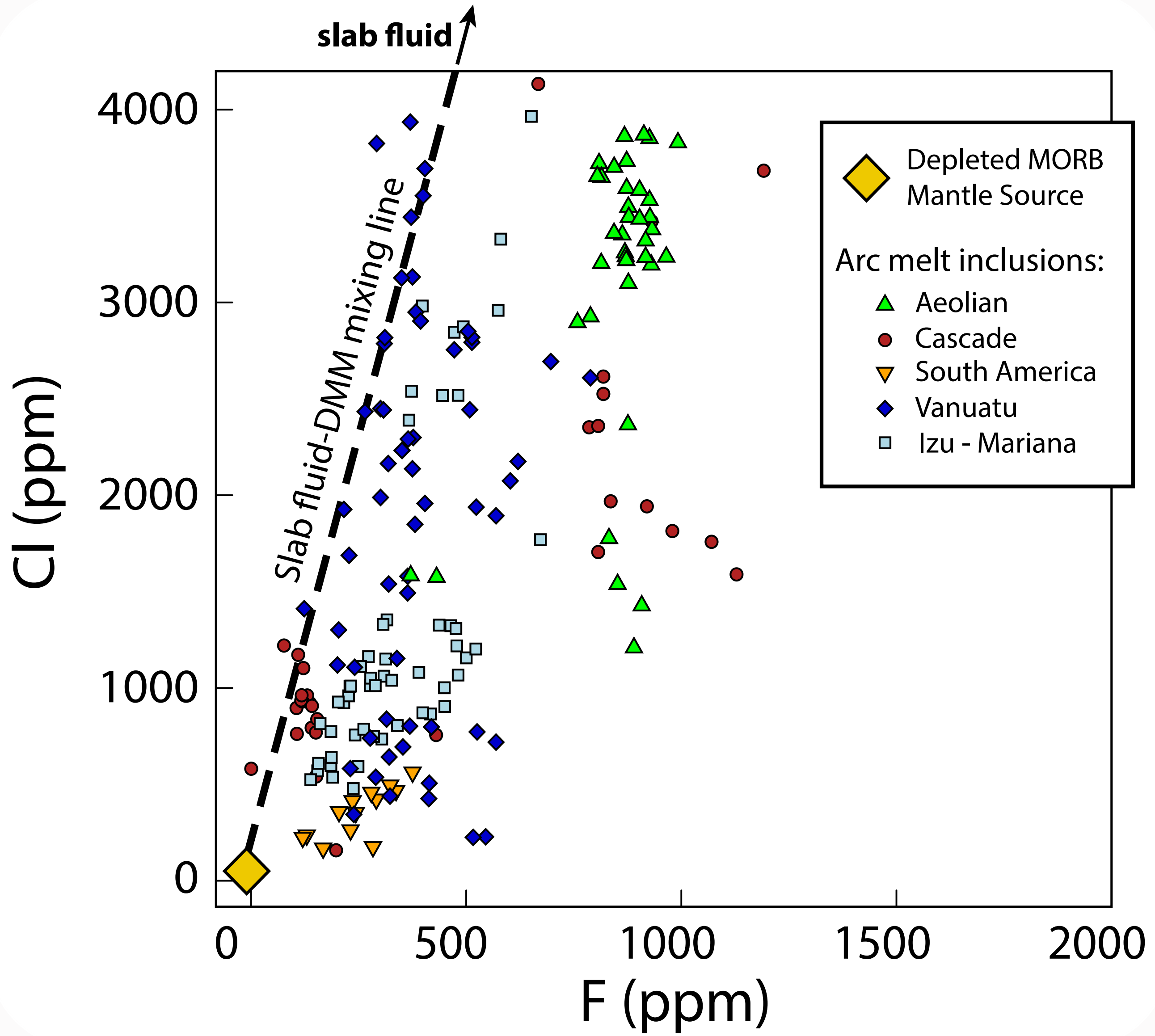

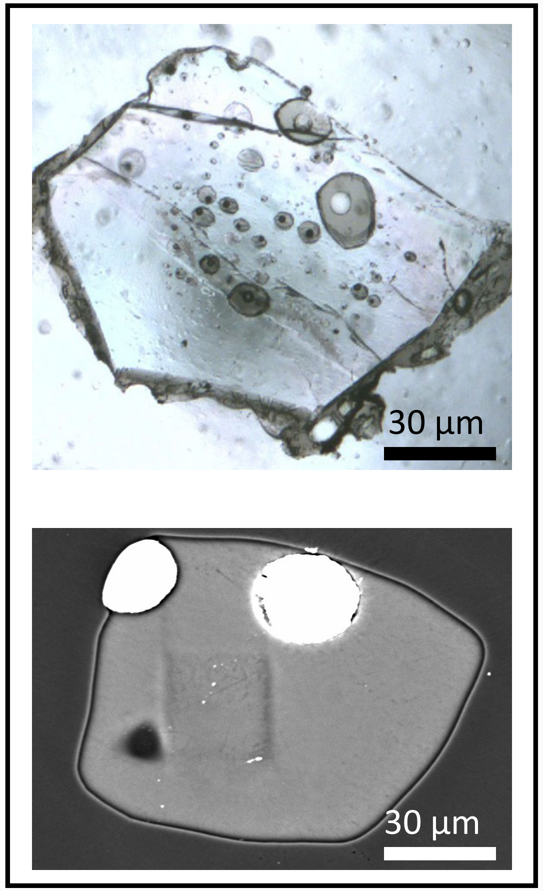

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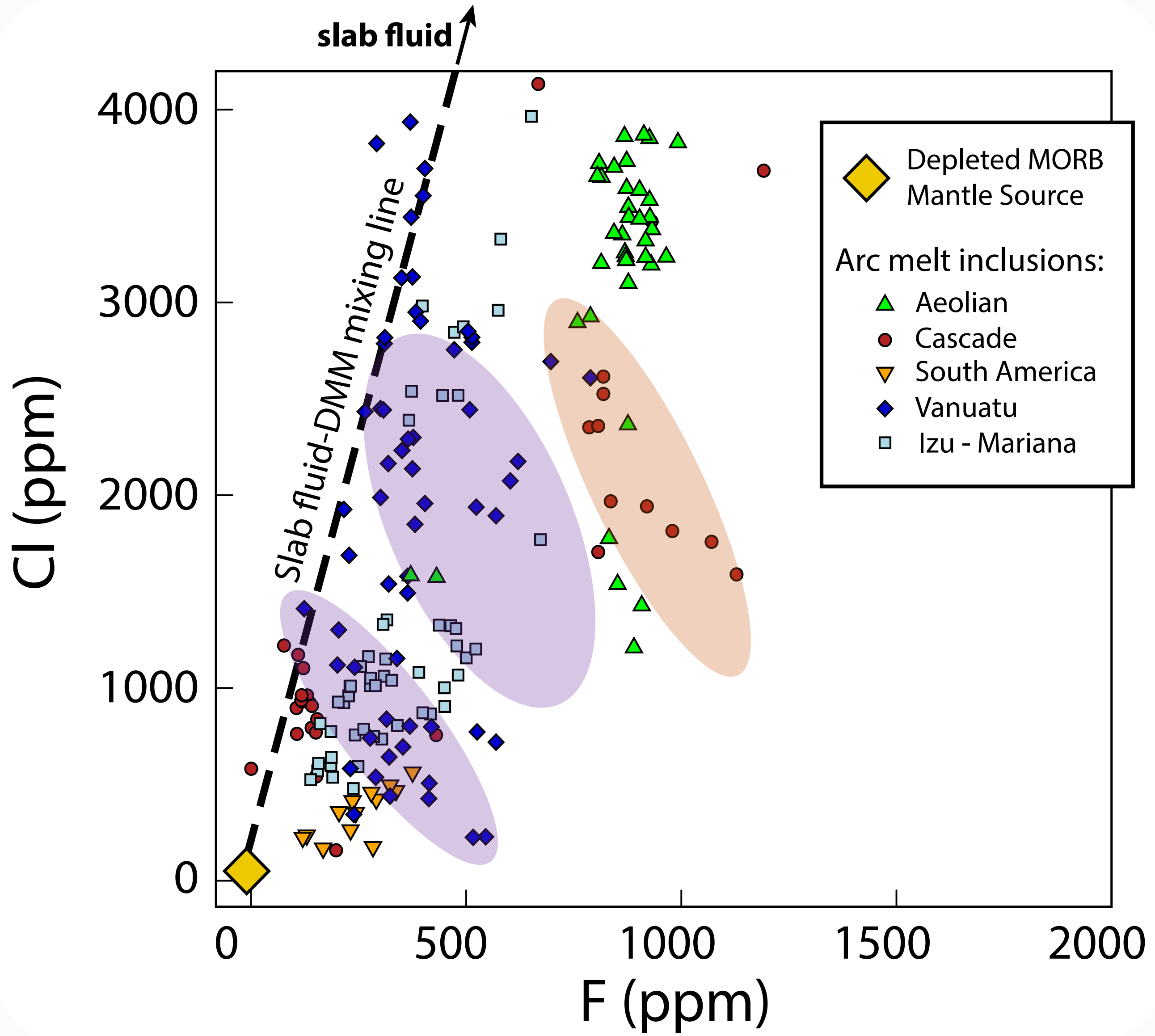

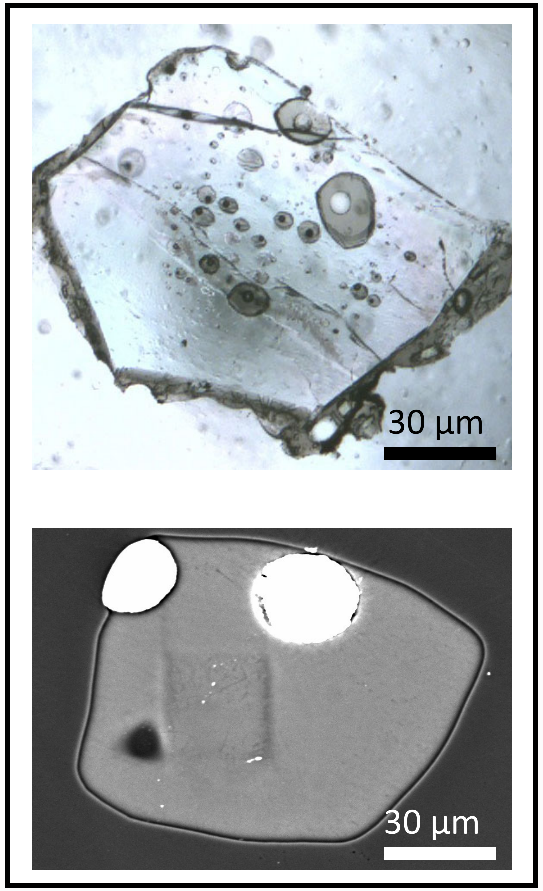

Photos: Marion Le Voyer

Main questions:

- What can we learn from the large range of F and Cl abundances in volcanic gases and melt inclusions ?

  - ➢ About the source of magmas ?

  - ➢ Behavior during hydrous melting ?

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  - ➢ About the source of magmas ?

  - ➢ Behavior during hydrous melting ?

  - ➢ Are they tracing the **source or a melting processes** in SZ ?

How to study volatile elements behavior ?

Starting materials

- 30:70 mixture of peridotite and basalt compositions (after Gaetani and Grove, 1998)
  between 0.2 to 5.9 wt% of water (dissolved in the melt)
  - **Olivine, Opx, Cpx**
  - **Amphibole, Opx**

  + basalt
- synthetic shoshonite (after Holbig and Grove, 2008)
  - **Phlogopite, Olivine**
  - **Phlogopite, Amphibole**

  + hydrous shoshonite

All doped with trace elements including Cl and F.

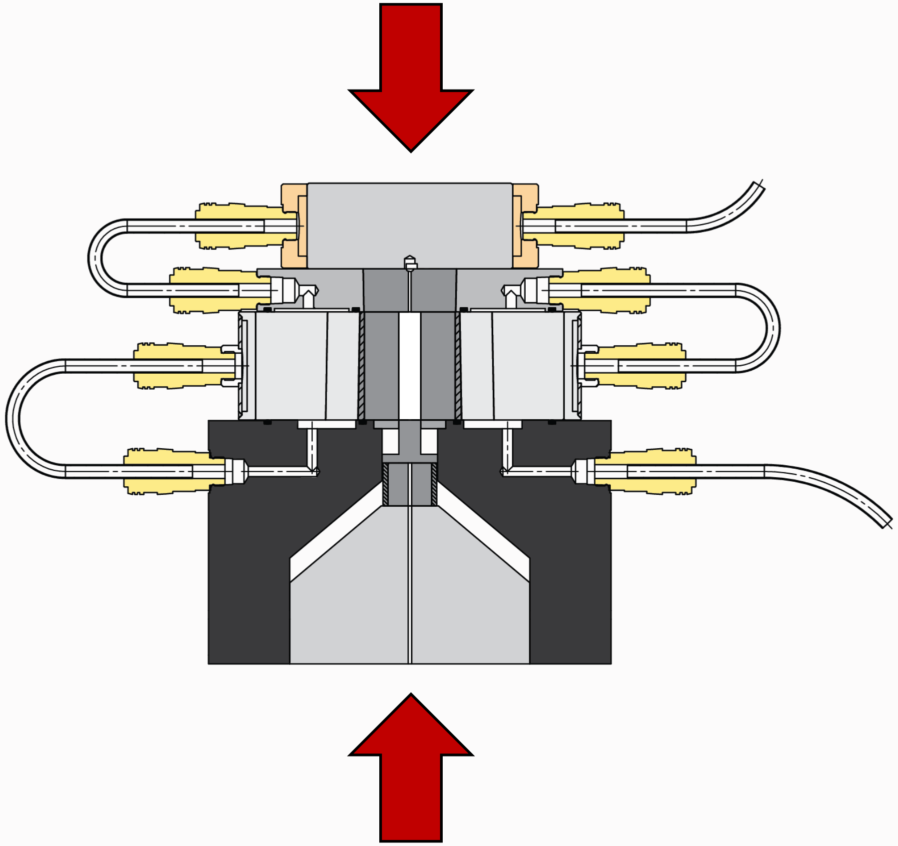
Experimental conditions

Conditions of the mantle wedge:

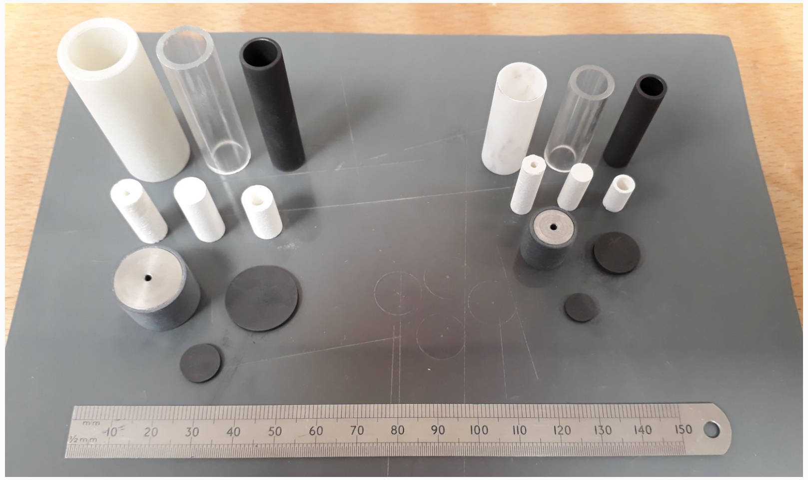
0.8 GPa - 2.5 GPa & 1040°C - 1430°C

Piston-cylinder principles

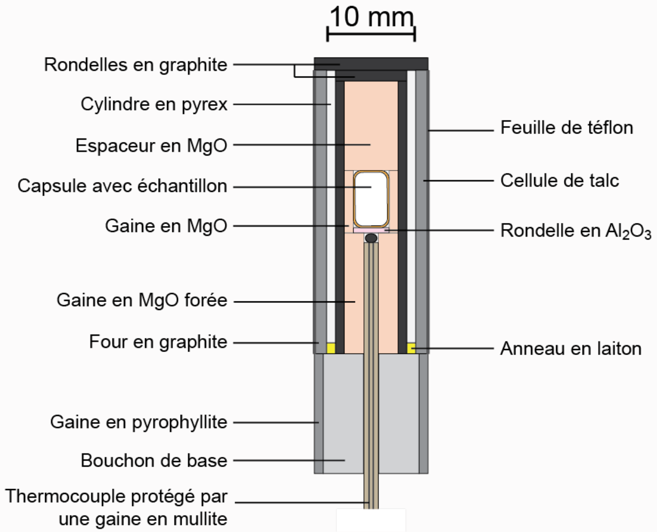
Counter pressure



Pressure



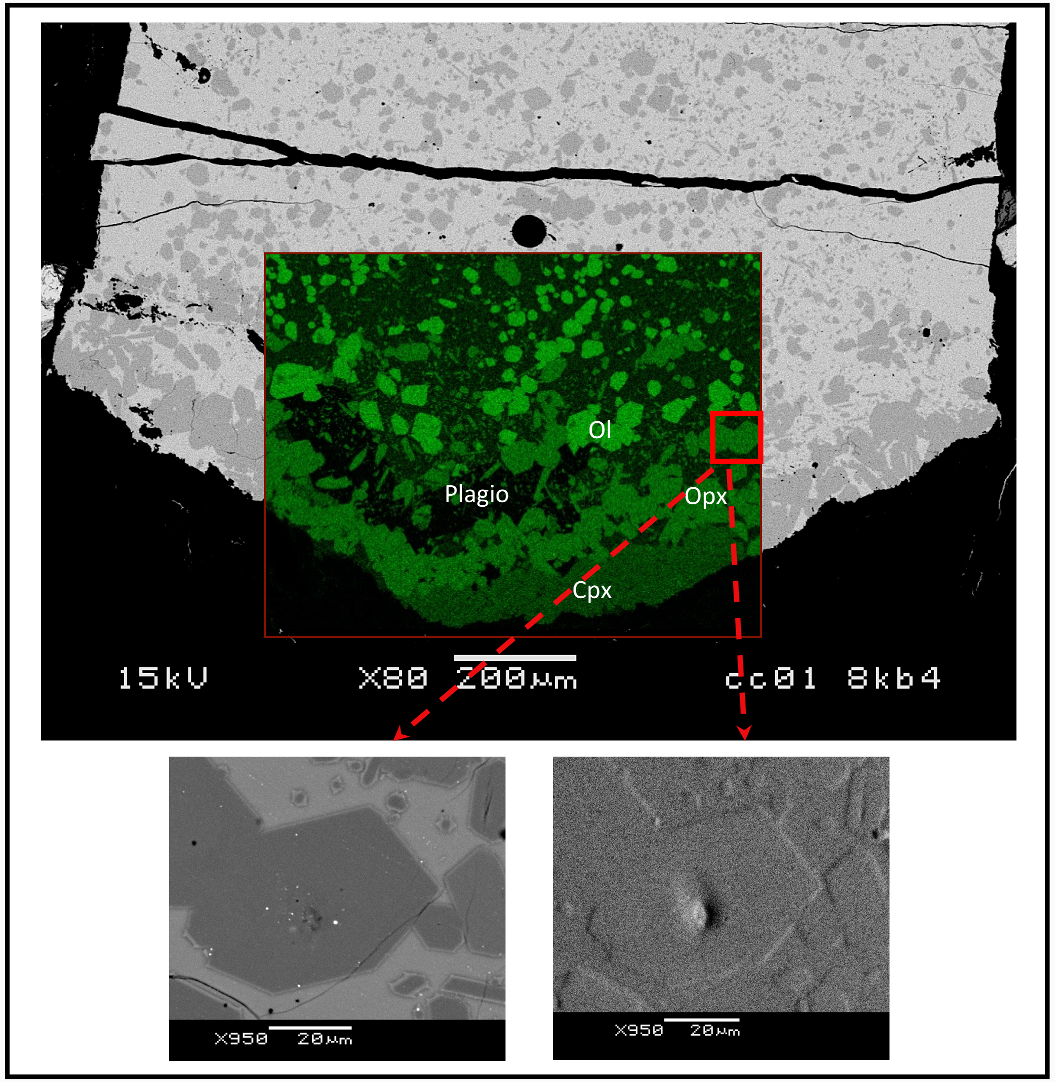
Assemblage 1/2”



How to study volatile elements behavior ?

3 types of experiments:

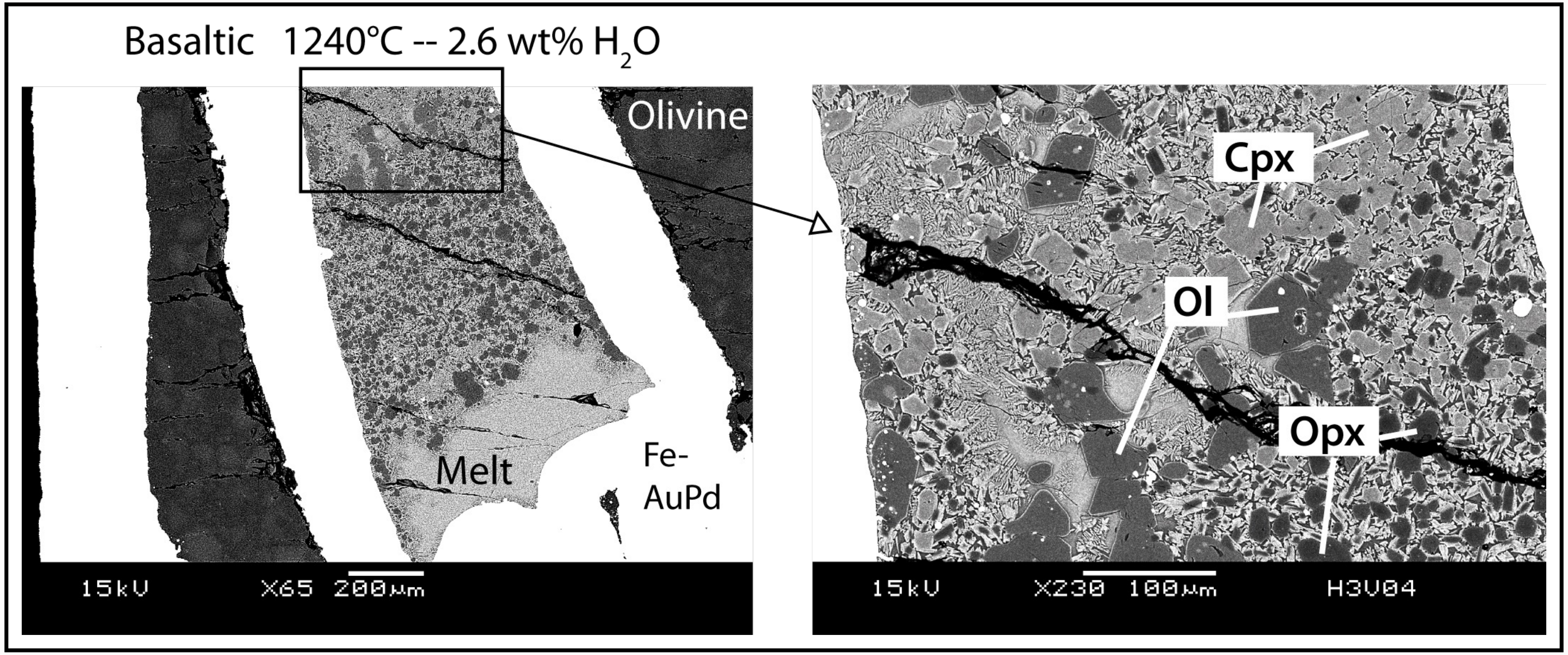
1 **Anhydrous** experiments



How to study volatile elements behavior ?

3 types of experiments:

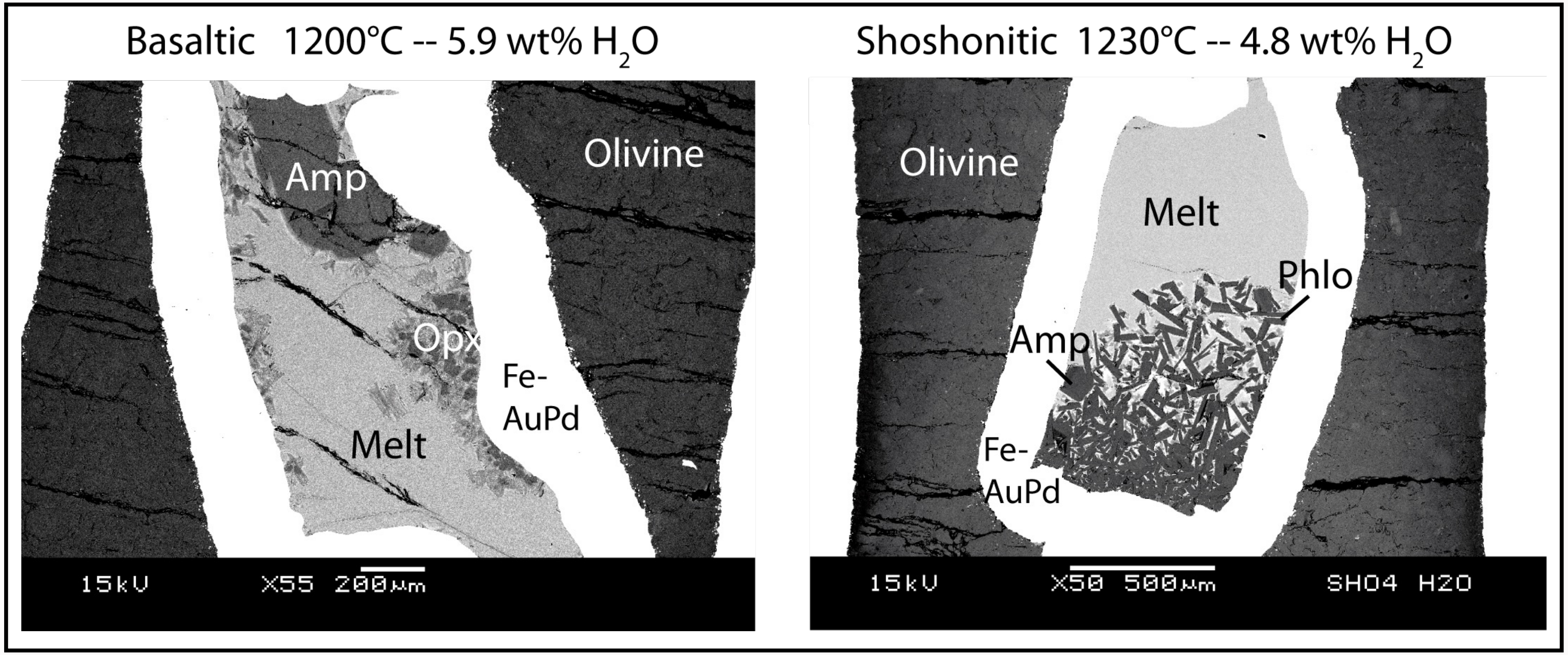
2 **Hydrous** experiments with **variable** H_2O doping.



How to study volatile elements behavior ?

3 types of experiments:

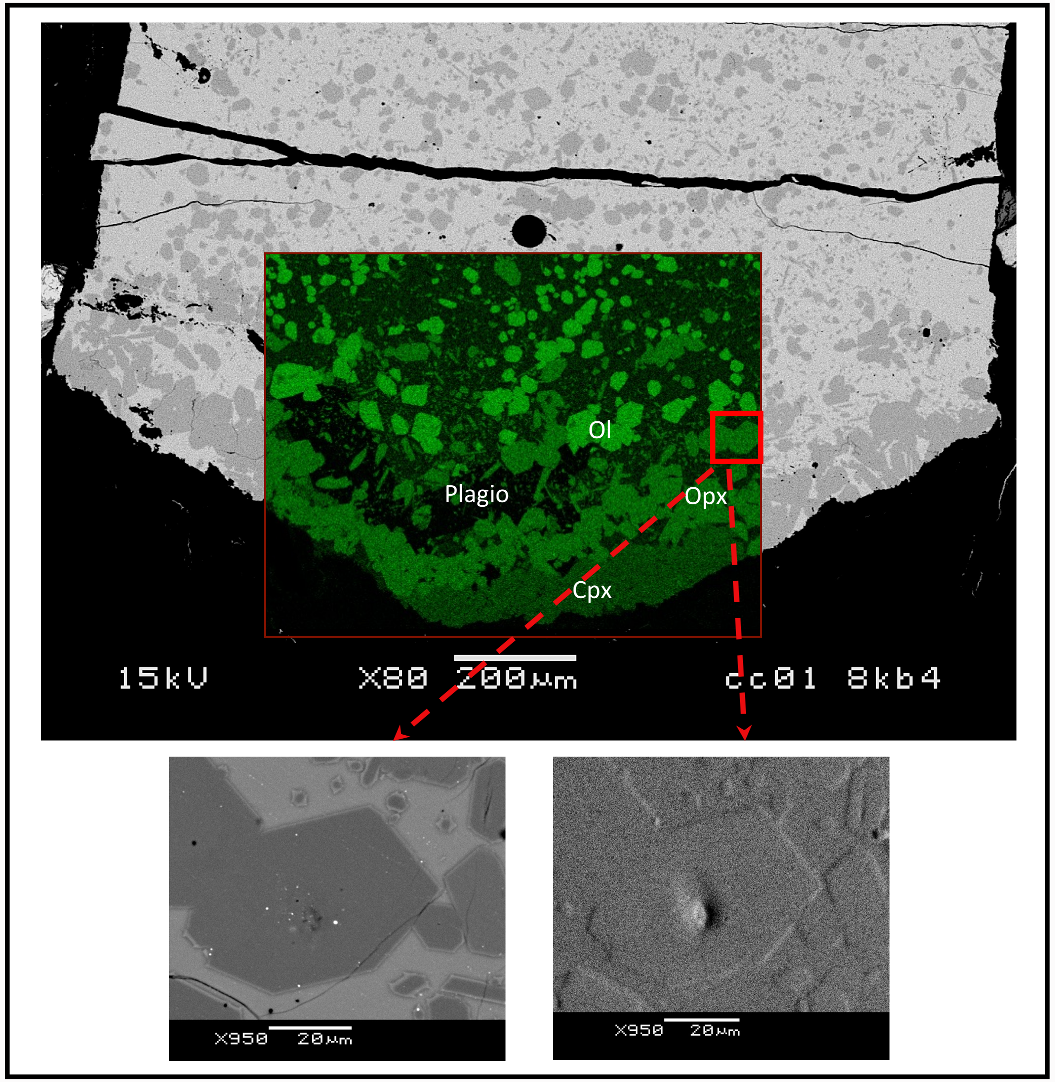
3 Experiments with **hydrous minerals**



How to study volatile elements behavior ?

Determination of F and Cl partition coefficients between minerals and silicate melt :

$$D_F^{\text{mineral-melt}} = \frac{C_F^{\text{mineral}}}{C_F^{\text{melt}}}$$



Modelling

- Fluid induced melting

$$C_i^{\text{arc magmas}} = \frac{C_i^{\text{DMM}}(1-X_f) + X_f * C_i^{\text{slab flux}}}{\boxed{\bar{D}_i^{\text{lherzolite/arc magma}}} + \chi\left(1-\boxed{\bar{P}_i^{\text{lherzolite/arc magma}}}\right)}$$

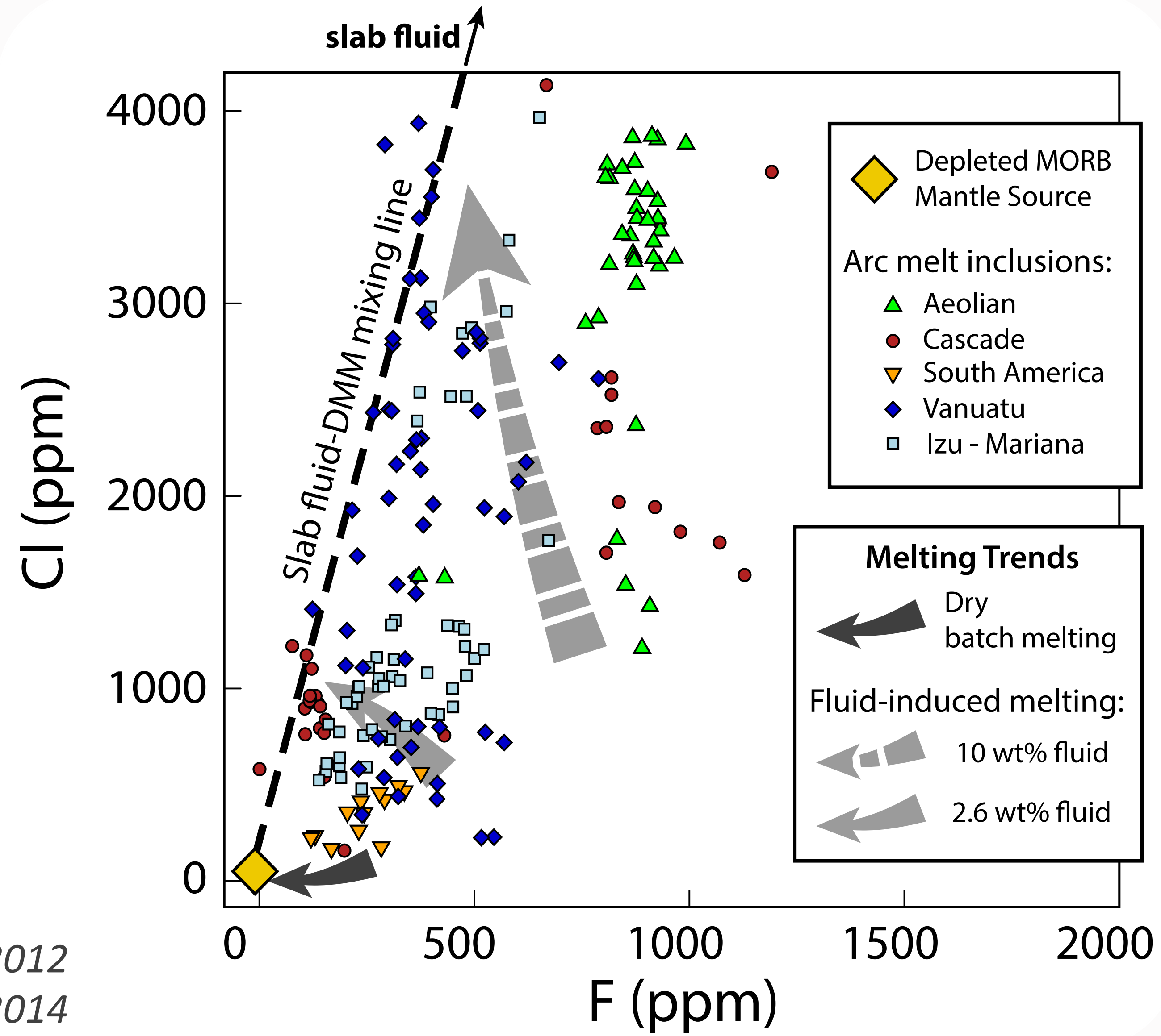
X_f = amount of fluid released by the slab

χ = degree of melting

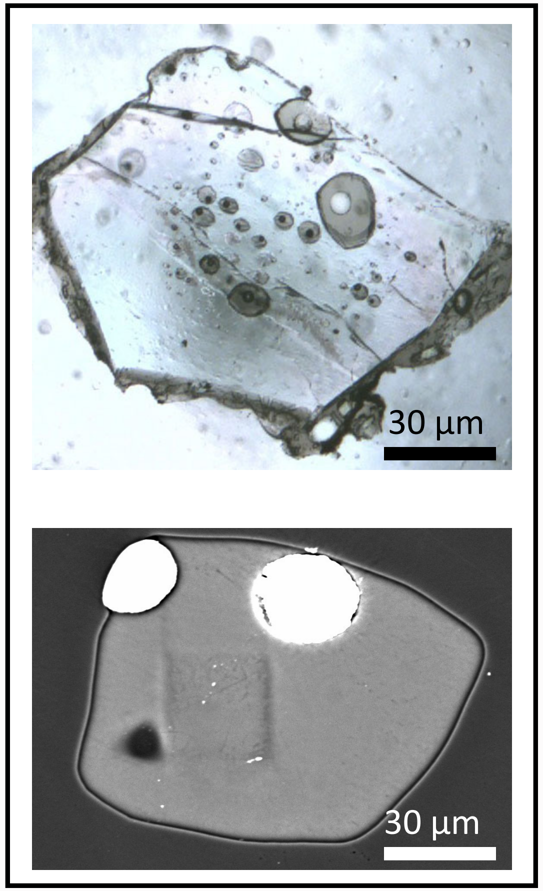
$\bar{D}_i^{\text{lherzolite/arc magma}}$ = mineral proportion weighted partition coefficients

$\bar{P}_i^{\text{lherzolite/arc magma}}$ = weighted partition coefficients
(0.62 Cpx + 0.51 Opx + 0.12 Sp = 1 Melt + 0.25 Ol)

Processes in subduction zones



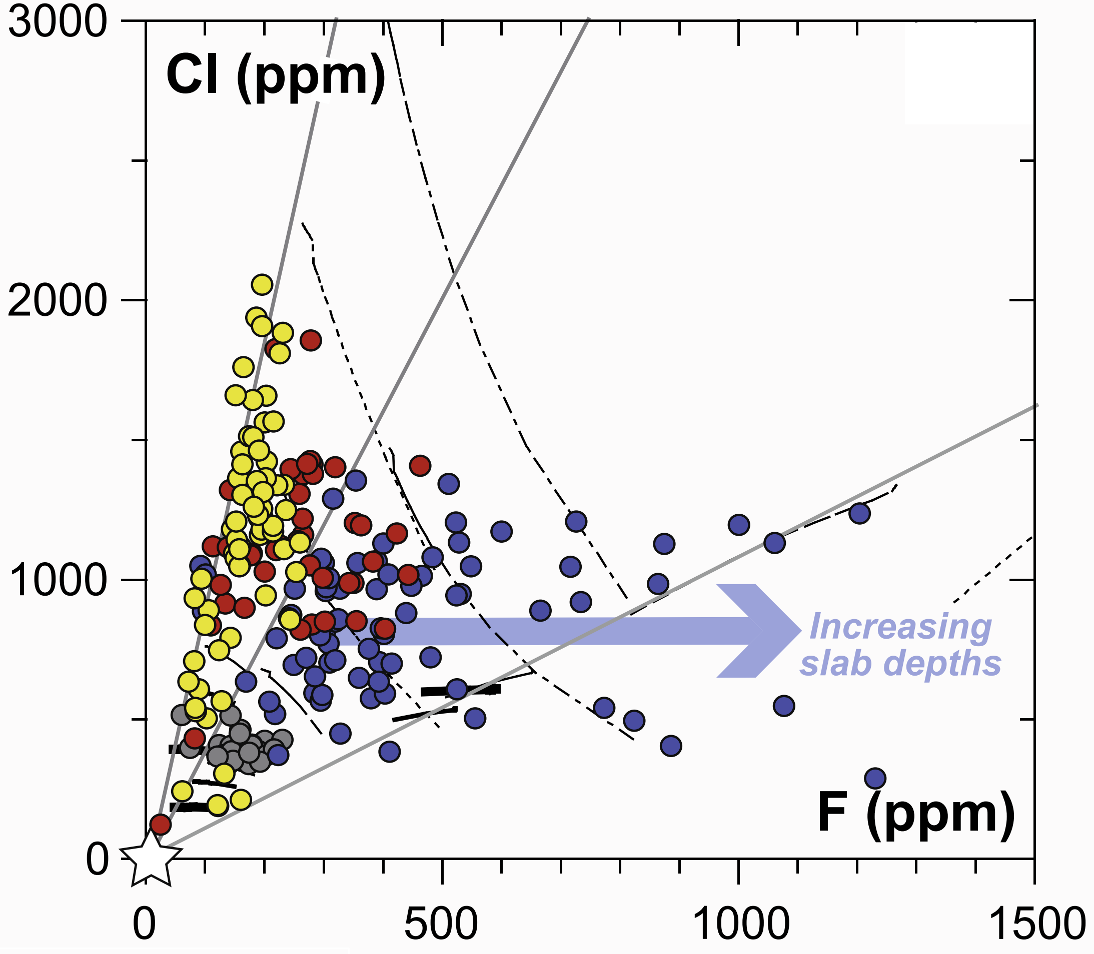
Dalou et al. 2012
Dalou et al. 2014



Photos: Marion Le Voyer

Processes in subduction zones

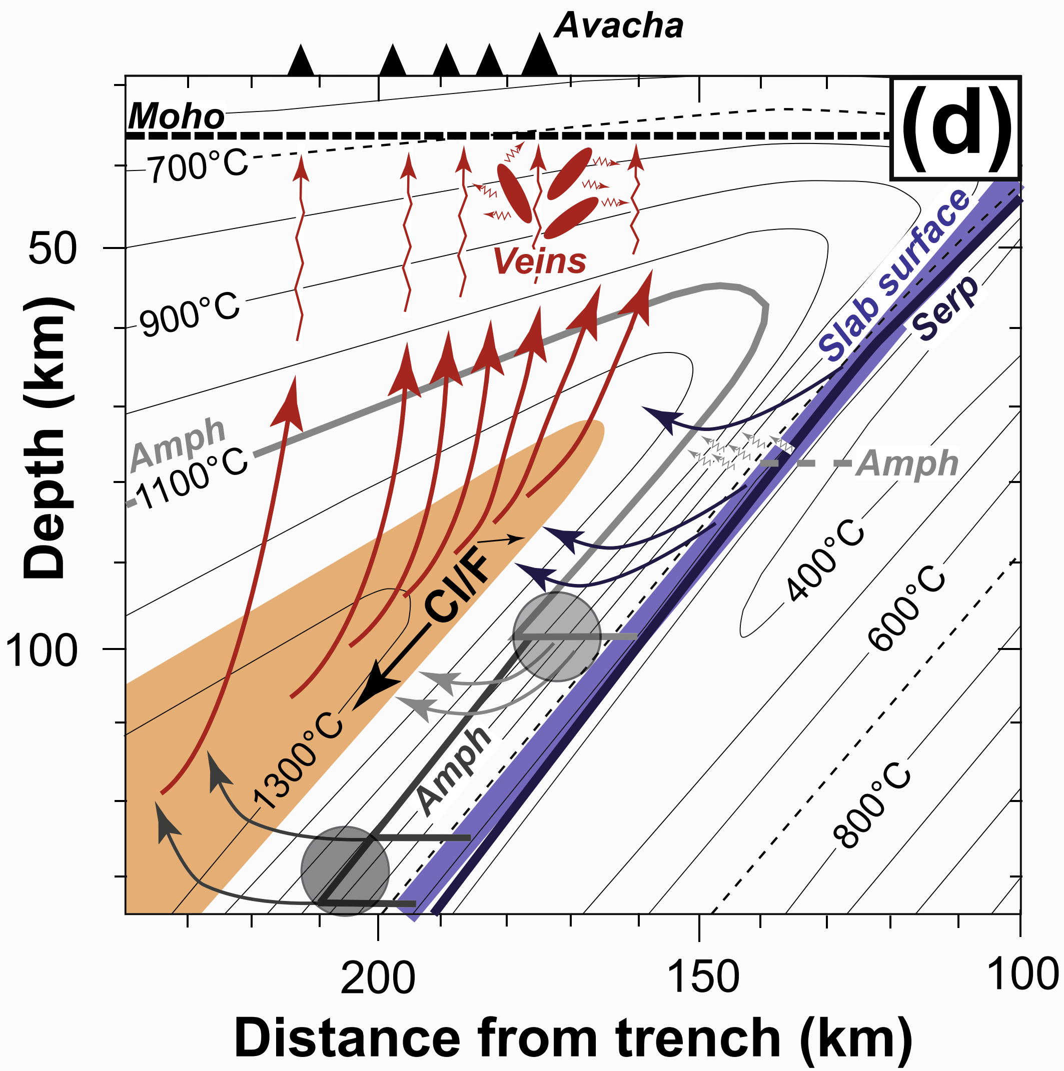
Kamchatka



Bénard et al. 2017

Processes in subduction zones

F and Cl trace the presence of amphibole or micas in the source of arc melt inclusions (not fluid-induced melting).



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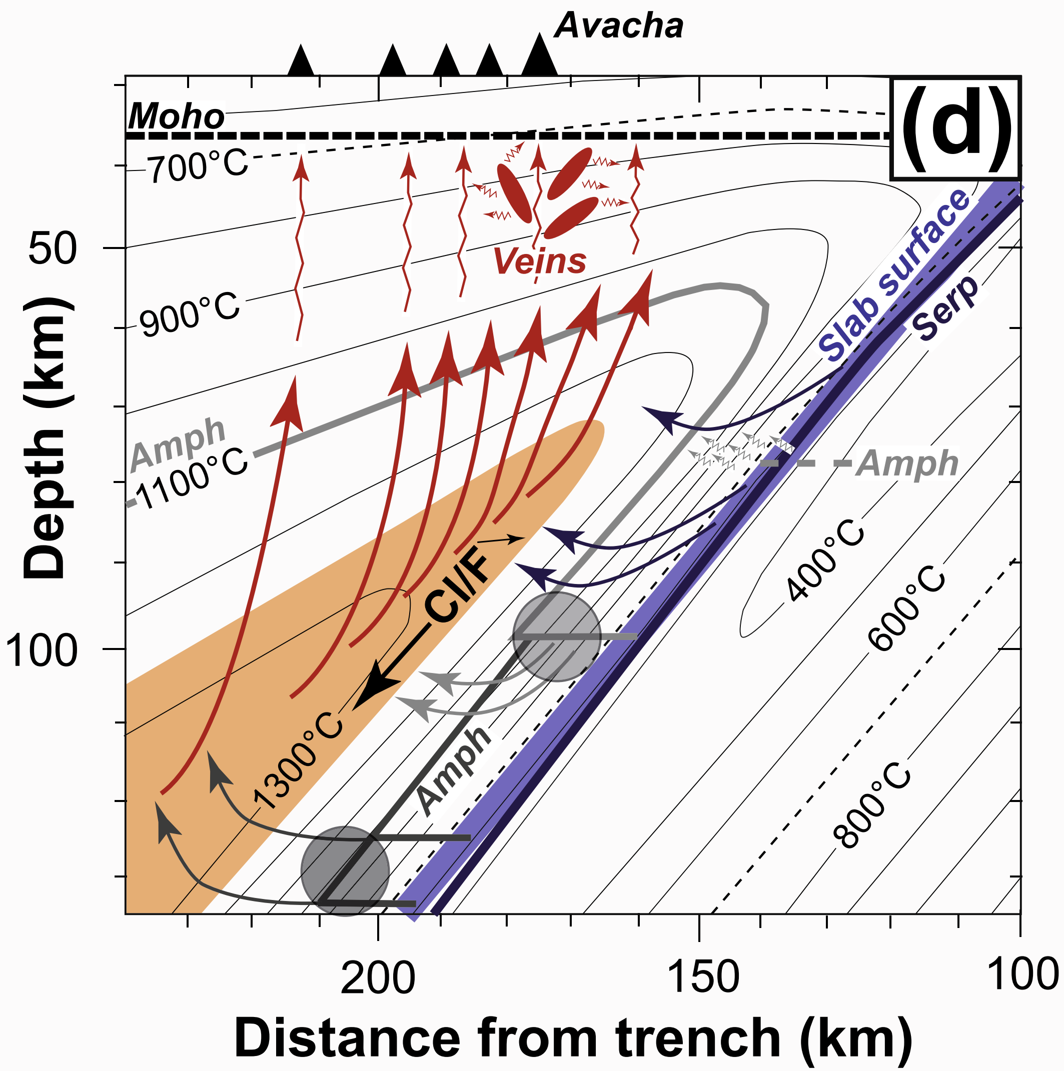

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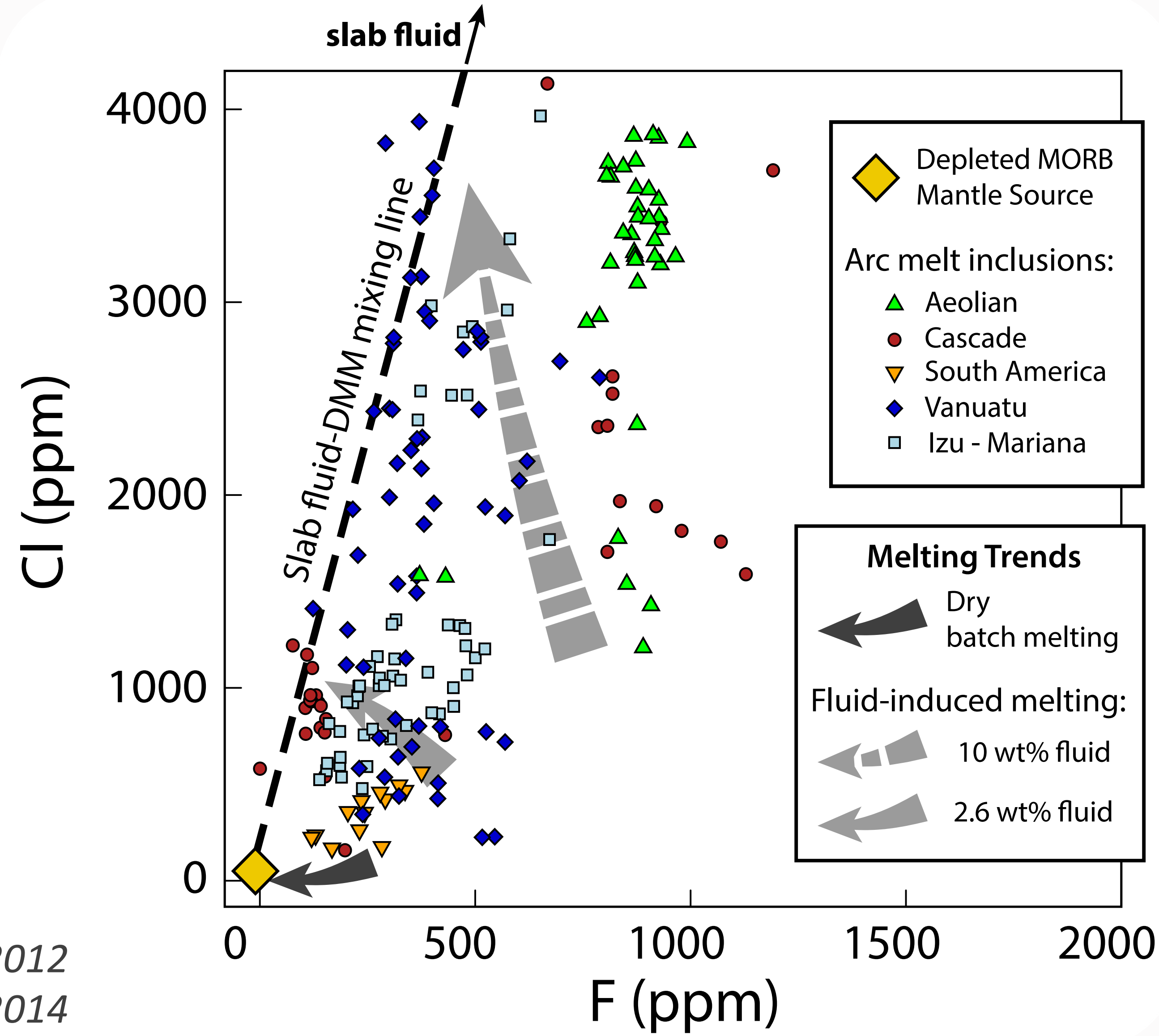

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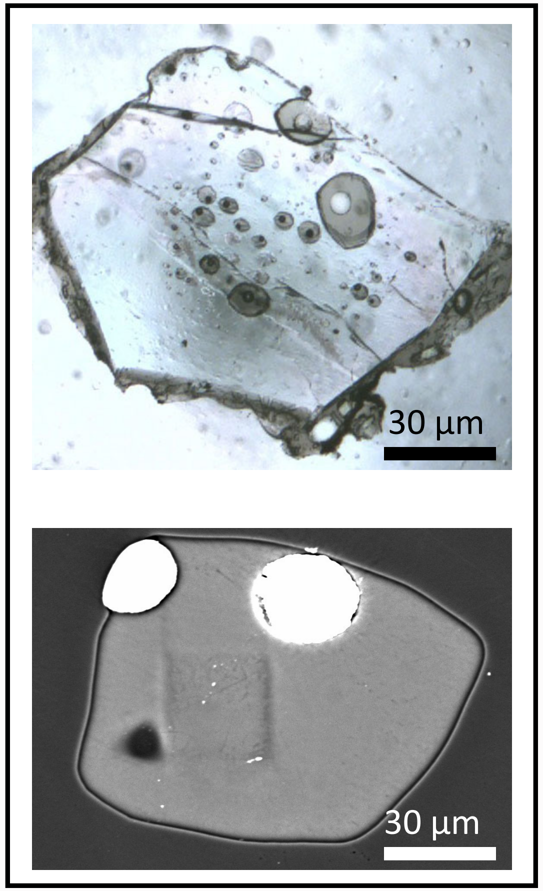
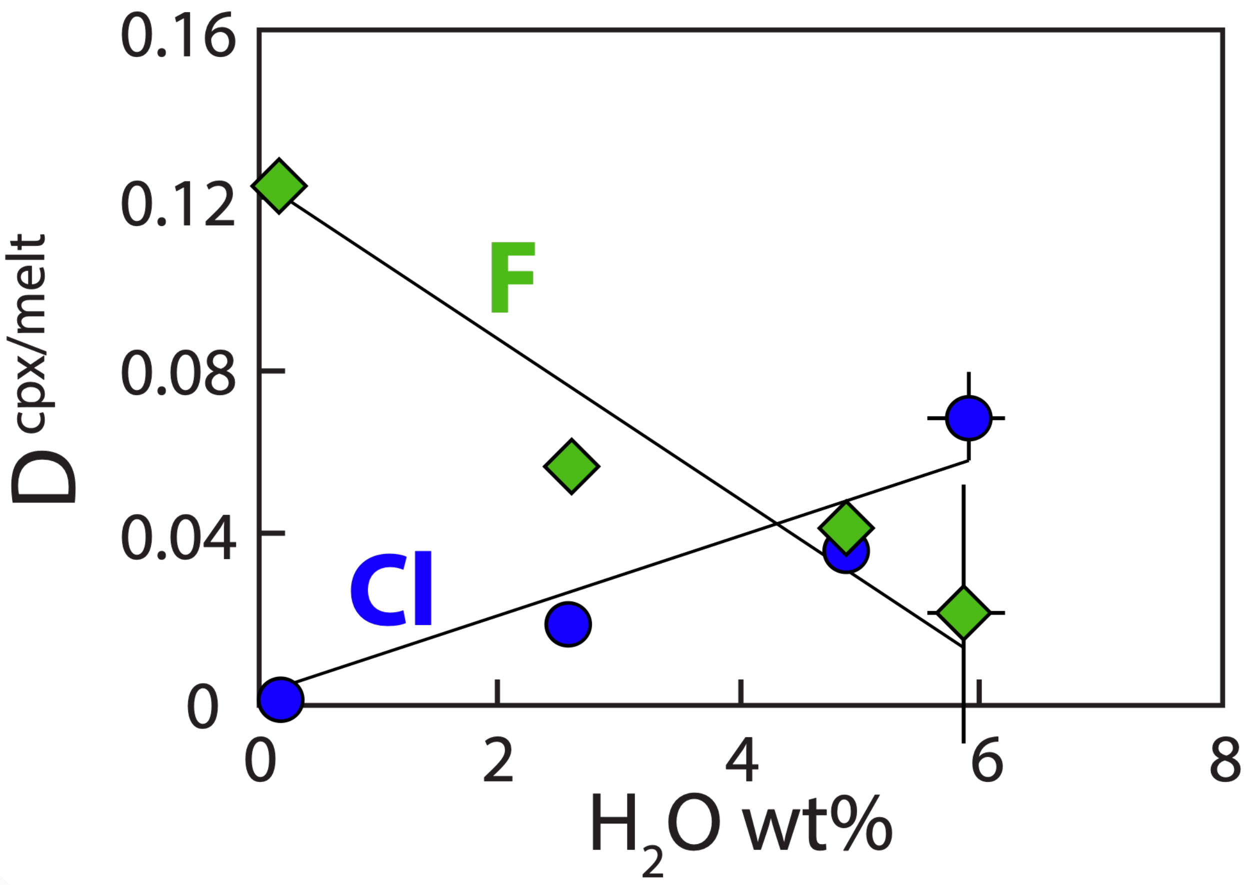


Photos: Marion Le Voyer

F and Cl fractionation



F and Cl partition coefficients with H_2O

F and Cl fractionate differently with H_2O

Why ?

Dalou et al. 2014

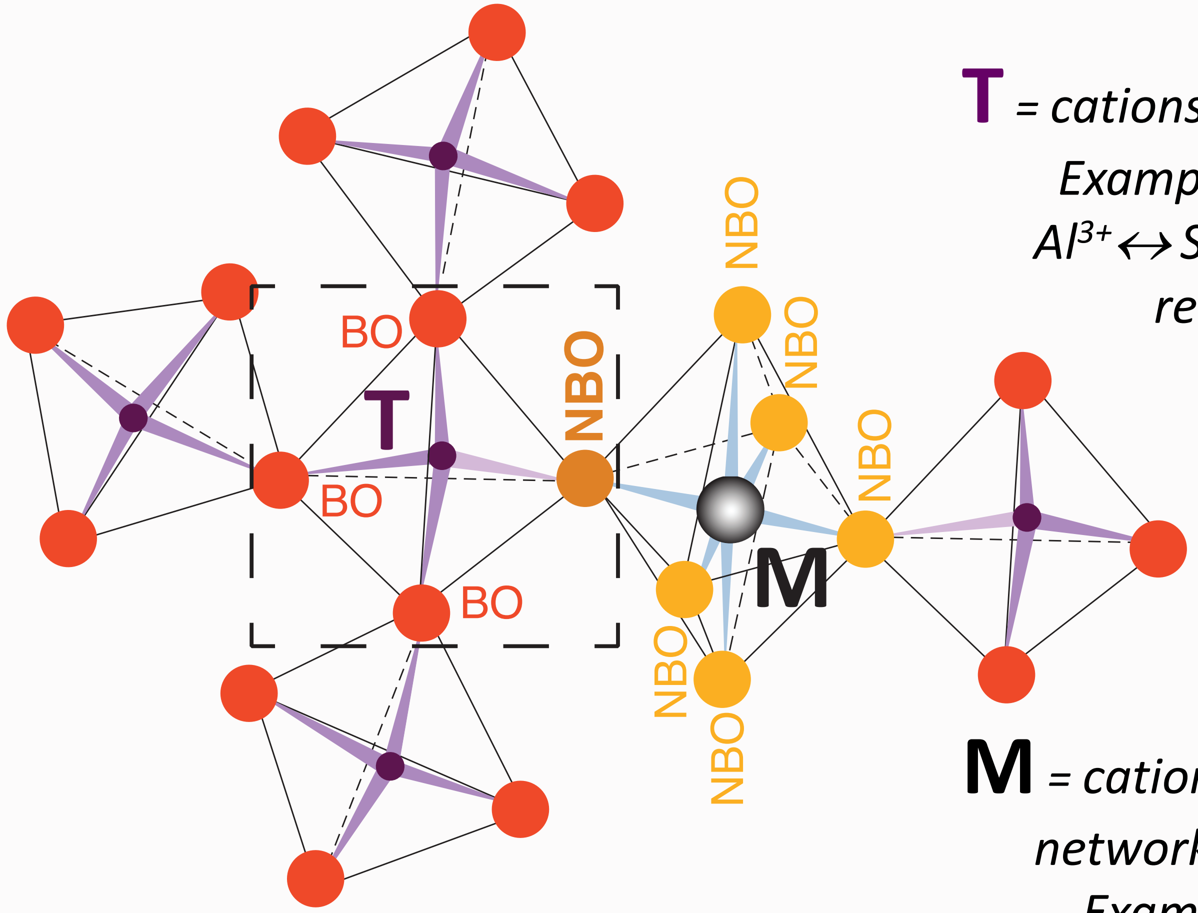
Why F and Cl behave differently with H_2O?

variation D_{Cl} and D_F with H_2O

Crystal chemistry variation with H_2O

Melt structure variation with H_2O
=
<u>depolymerization</u>

Melt structure



T = *cations in tetrahedral coordination*

Example : Si^{4+}, Al^{3+}

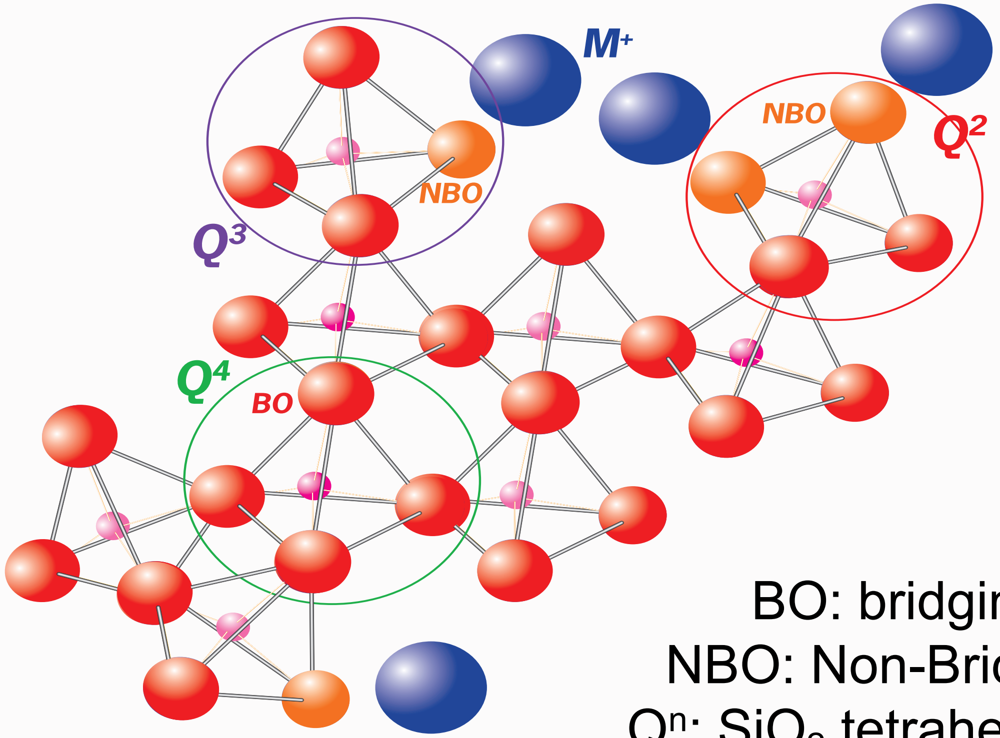
$Al^{3+} \leftrightarrow Si^{4+}$ substitution requires charge balance

M = *cations in octahedral coordination*

network-modifiers or charge balance

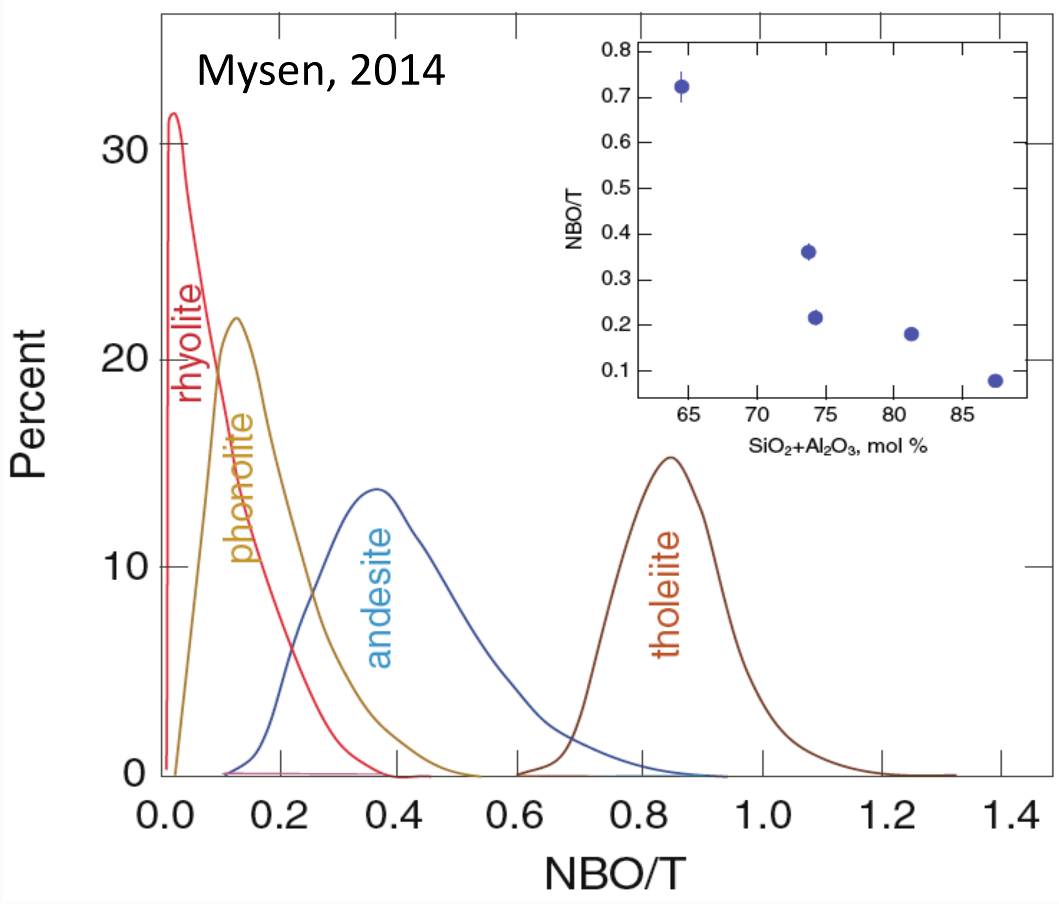
Example: Na^{+}, K^{+}, Ca^{2+}…

Melt structure



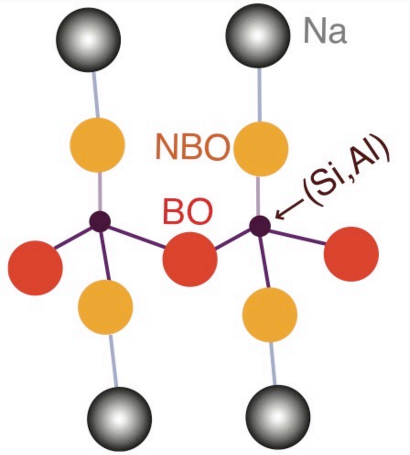
BO: bridging oxygen
NBO: Non-Bridging Oxygen
Q^n: SiO_2 tetrahedron with n BO

NBO/T: number of NBO per tetrahedron, a quantification of the polymerization of silicate melts…

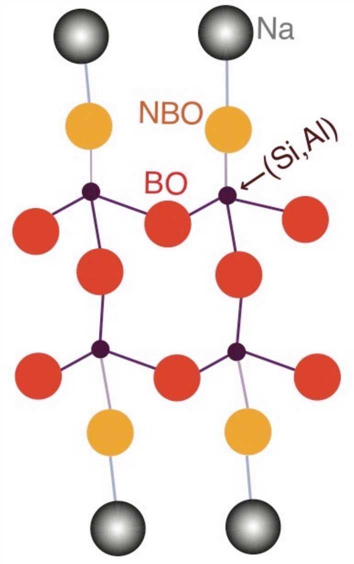


Qs species present in a melt

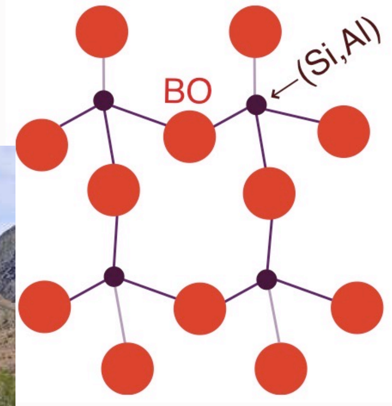
Q2



Q3



Q4



Depolymerization

= increase of NBO/T

= melt becomes less viscous

How to study F and Cl solubility

Simple system along the join

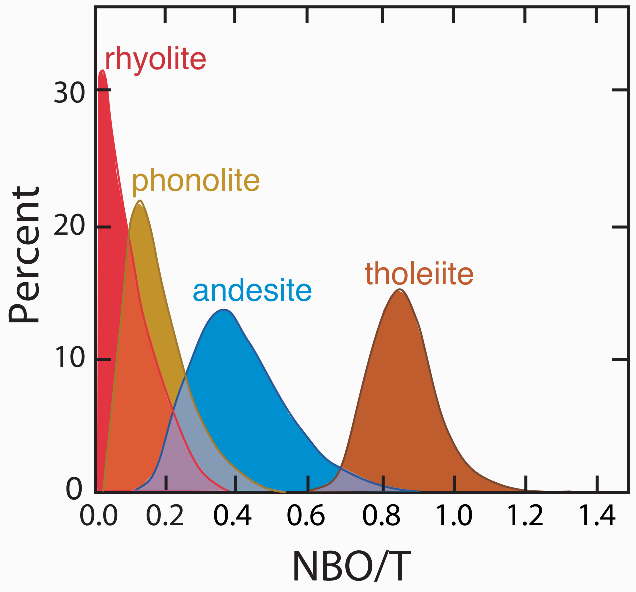
$Na_2Si_3O_7$-$Na_2(NaAl)_3O_7$

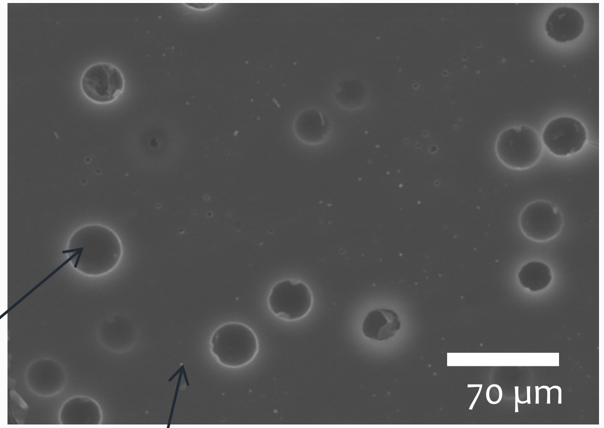
F and Cl as PdF_2 & $AgCl_2$

10 wt% of F & 5 wt% of Cl

Saturation of F and Cl

= presence of bubbles in the glass



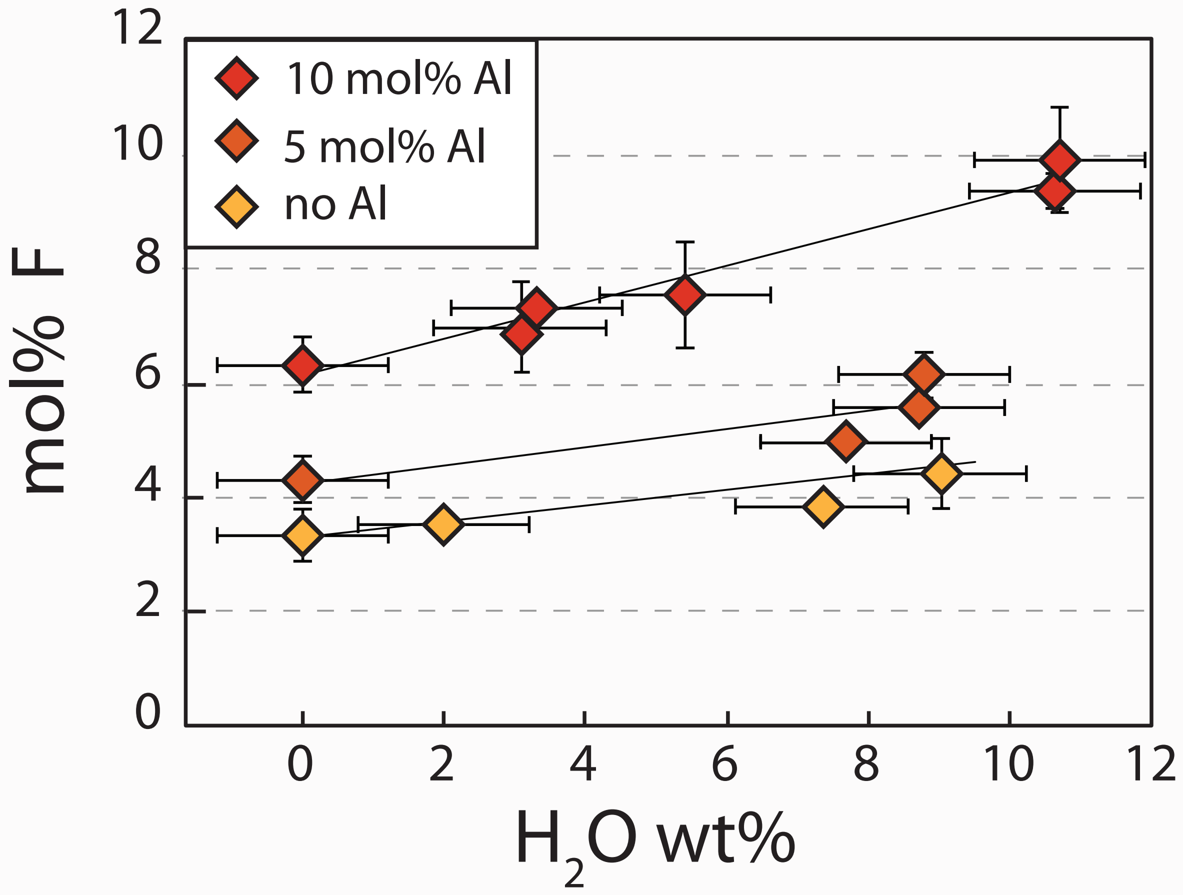


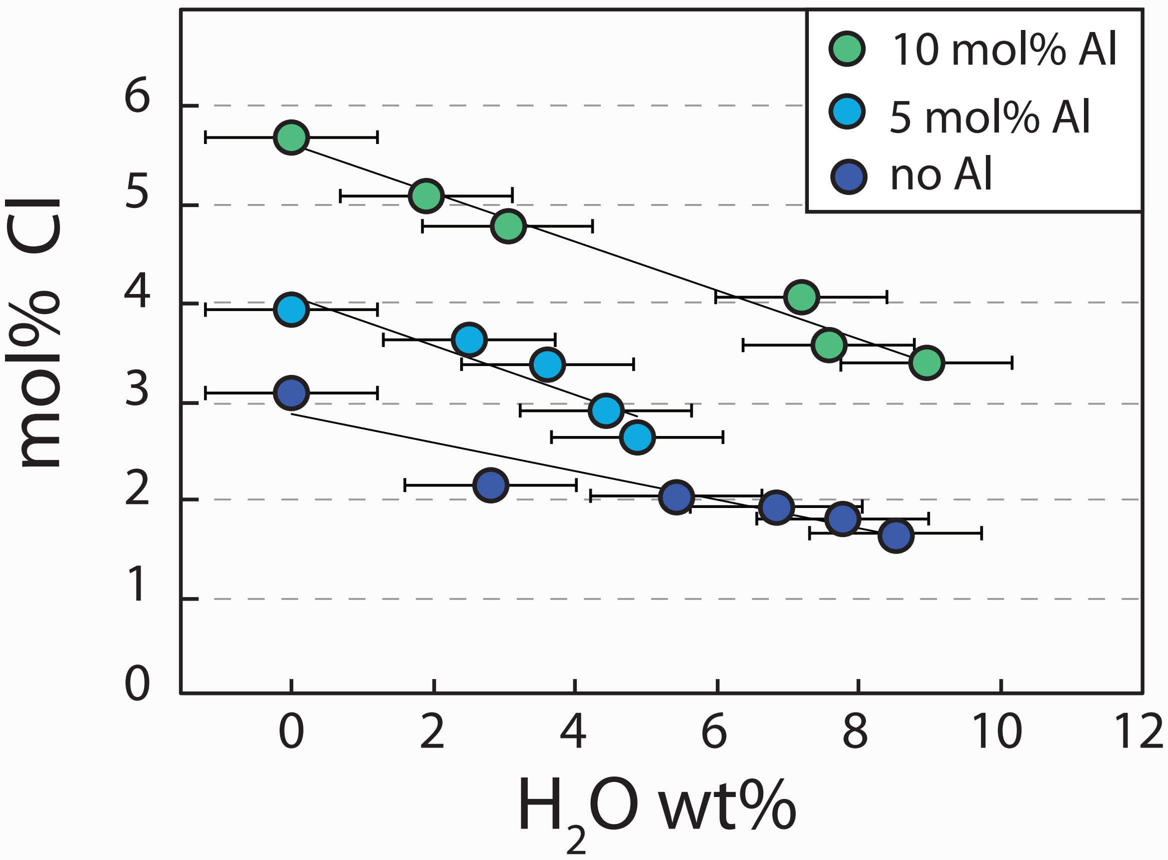
<u>Hydrous experiments:</u>

Up to 10 wt% (undersaturated in H_2O) as liquid H_2O

At 1.5 GPa – 1400ºC on piston-cylinder

The effect of H2O on F and Cl solubility

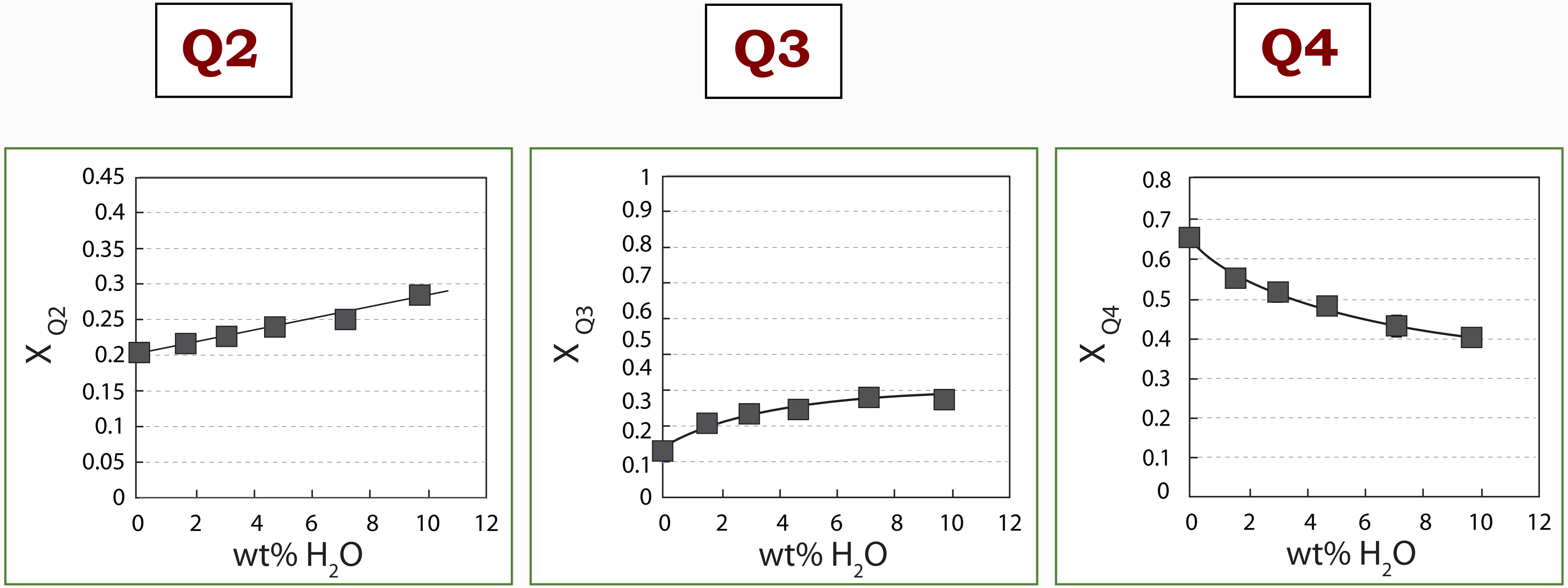




H_2O enhances F dissolution in silicate melt, while decreases Cl solubility.

In hydrous melts: variation of Qs

Effect of H_2O without F and Cl in Al-bearing melts:



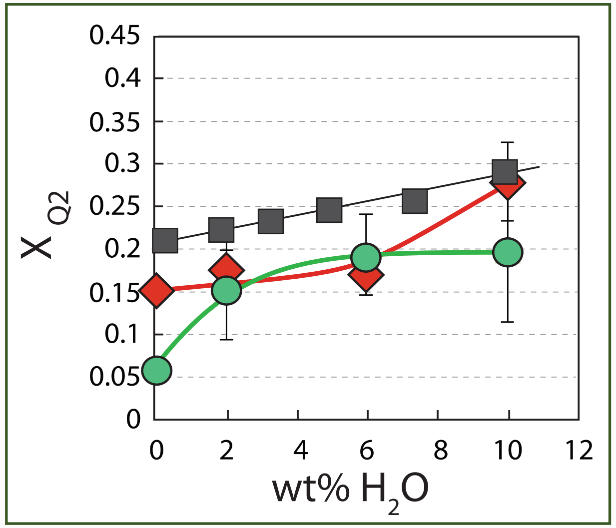
With H_2O: $Q4 + 2H_2O \longleftrightarrow Q2$

Modified from Mysen, 2005

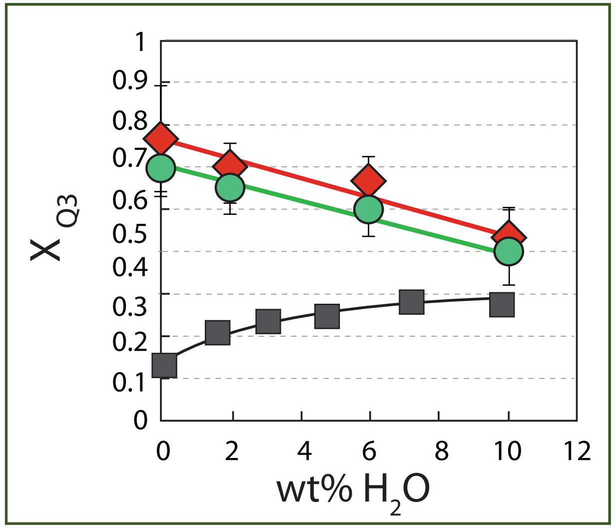
In hydrous melts: variation of Qs with F and Cl

With F ◆ and Cl ● in Al-bearing melts:

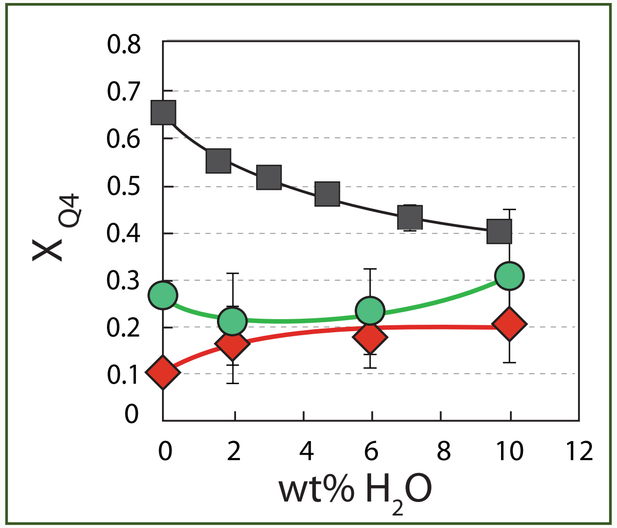
Q2



Q3

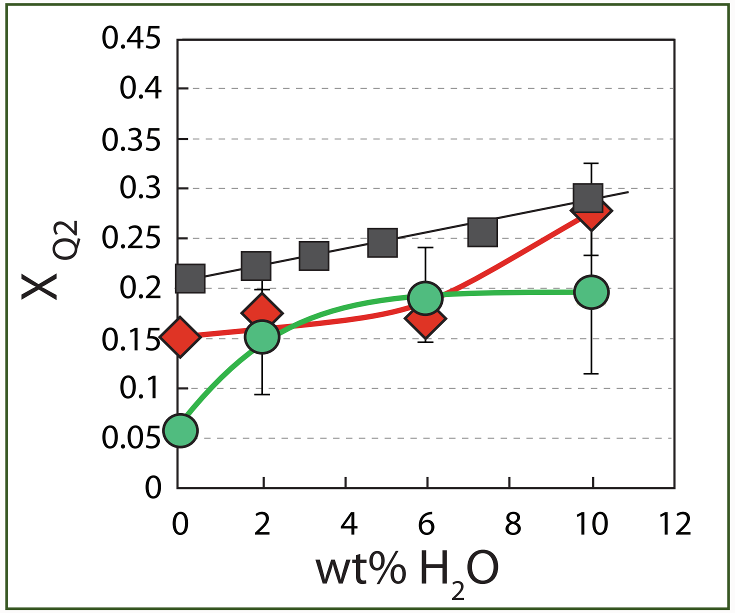


Q4



With H_2O + F or Cl: $Q3 + H_2O + F \text{ or } Cl \longleftrightarrow Q2$

Competition between H_2O and F or Cl



- less Q2 production

  ⟹ F and Cl decrease the depolymerizing effect of H_2O

  ⟹ Competition H_2O versus F or Cl

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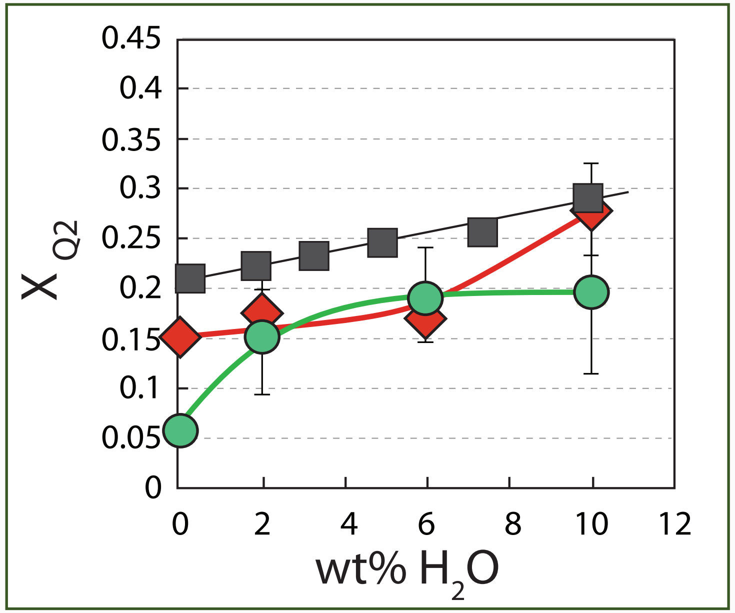

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- Since Mysen & Virgo (1986), we know that, in aluminosilicate melts, H_2O forms:

$Si(OH)_4$ ⇨ **Si-F ?**

Competition between H_2O and F or Cl

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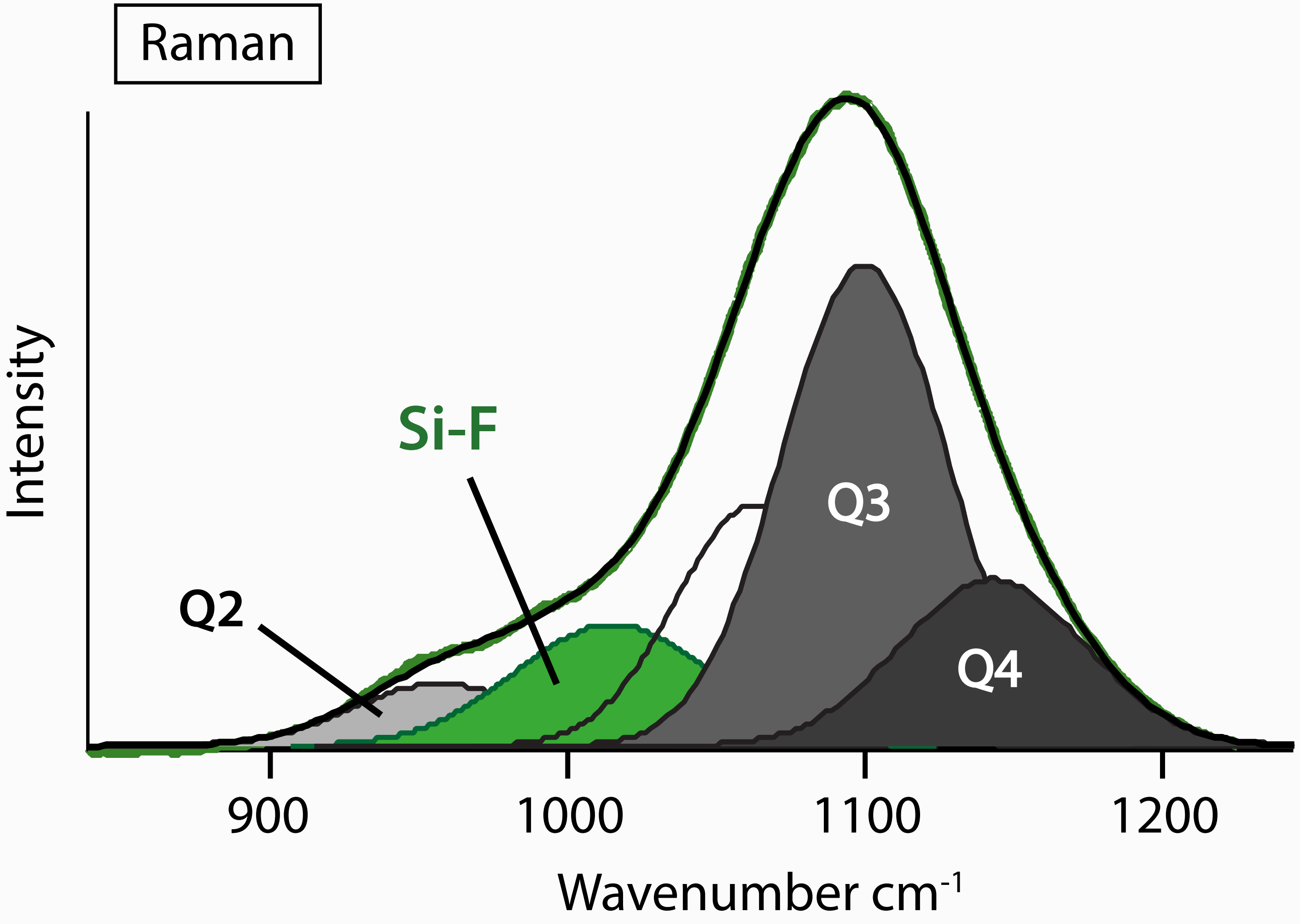

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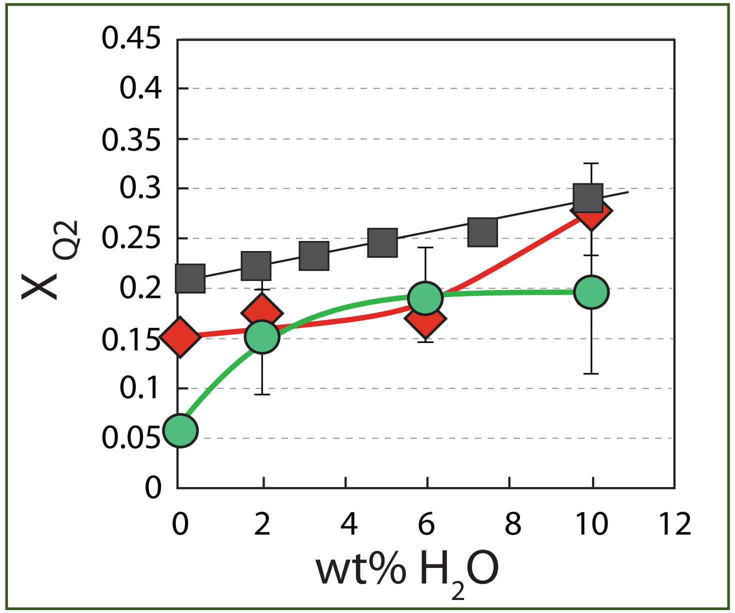

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$Si(OH)_4$ ⇨ **Si-F**

NaOH ⇨ **NaCl**

The effect of H_2O on F and Cl complexes

From solubility data and Qs species variation, we assume that the results of H_2O vs F or Cl competition is:

$3Q^4 + H_2O + F_2 = Q^3(F) + Q^2(H) + 1/2\ O_2$ ➡ $3\ SiO_2 + \mathbf{H_2O} + F_2 = \mathbf{Si_2O_3F_2} + SiO_3H_2 + 1/2\ O_2$

F solubility increases with H_2O

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F solubility increases with H_2O

As H_2O dissolved, Na become less concentrated in melts…

Cl solubility decreases with H_2O

In magmas

F becomes **more** solube with the increase of H_2O

→ F becomes **more** incompatible

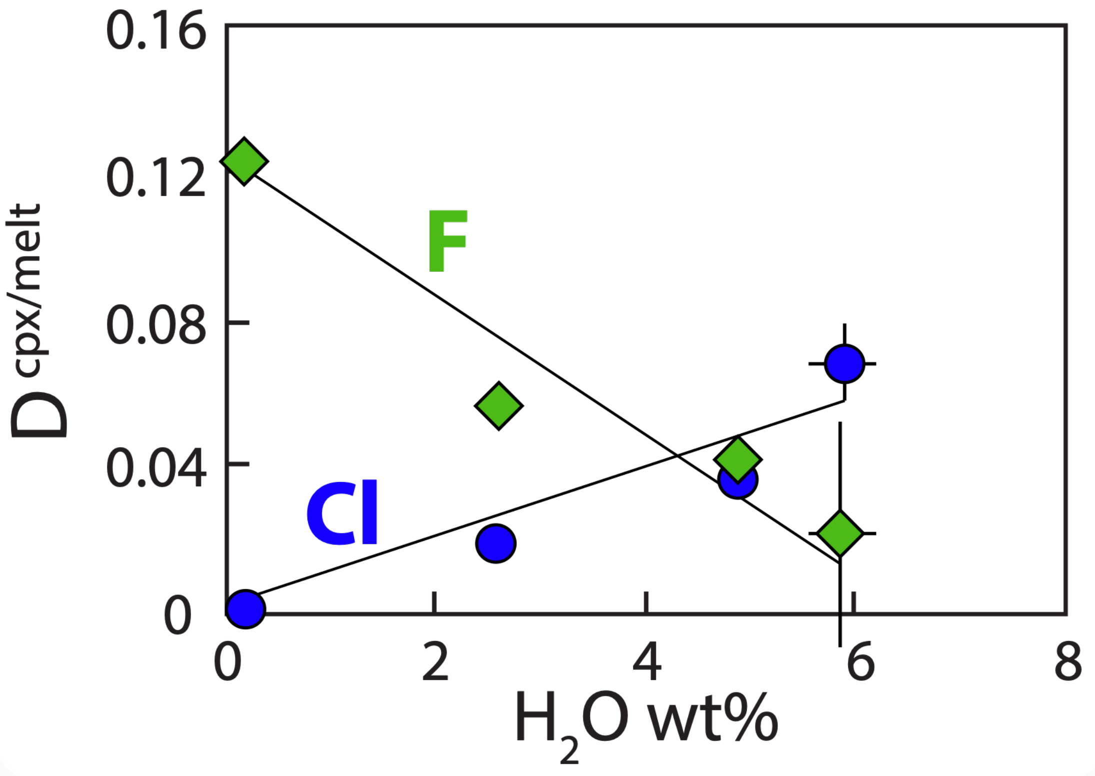
WHILE

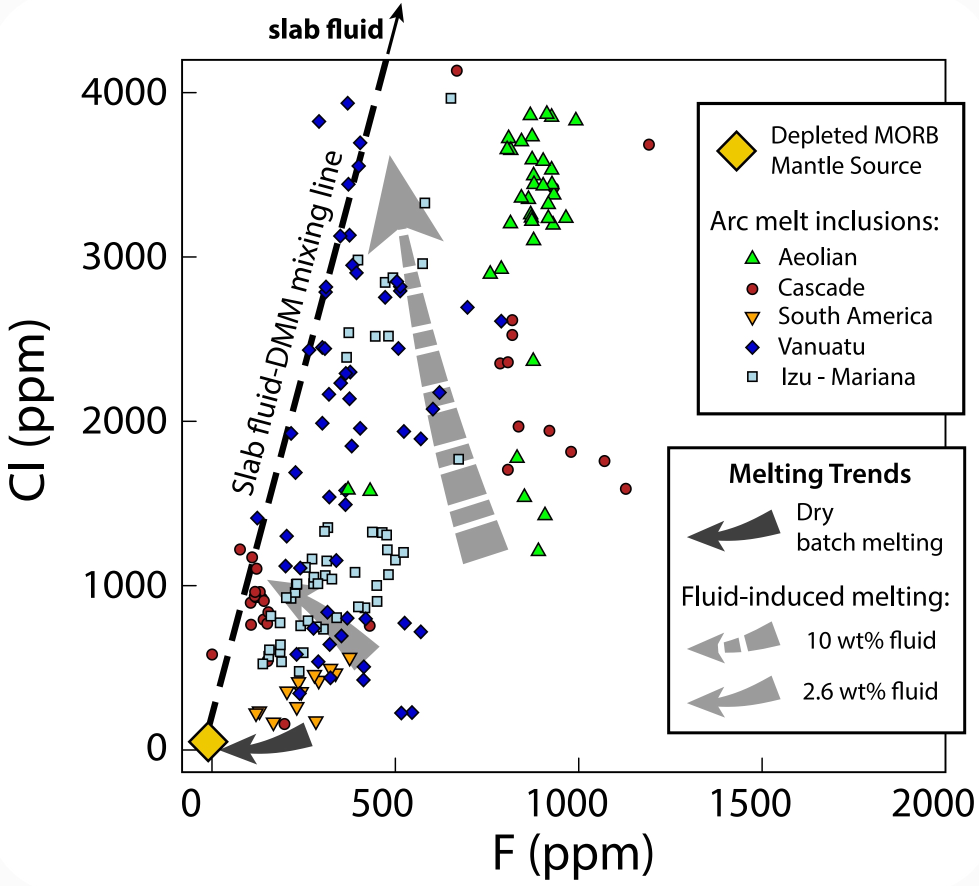
Cl becomes **less** soluble with the increase of H_2O

→ Cl becomes **less** incompatible

↳ Variable Cl/F ratios depending on the amount of H_2O dissolved in magmas

In magmas





Variable Cl/F ratios depending on the amount of H_2O dissolved in magmas

Remaining question:

- Why F and Cl are way less degassed than H_2O, CO_2 and S ?

	Convergent Plate[b]	Divergent Plate	Hot Spot
T(°C)	768	1130	1140
log fO_2	−14.41	−9.31	−8.82
H_2O	91.9	75.1	75.7
CO_2	4.6	13.1	3.2
H_2	0.5	1.59	0.95
H_2S	0.67	1.01	0.16
SO_2	1.44	7.84	19.4
HCl	0.76	0.42	0.17
HF	0.061	0.42[b]	0.18
CO	0.03	0.6	0.09

[a]Compiled from *Gerlach* [2004].
[b]Based on the proposed ratio HCl/HF = 1 by *Symonds et al.* [1994].

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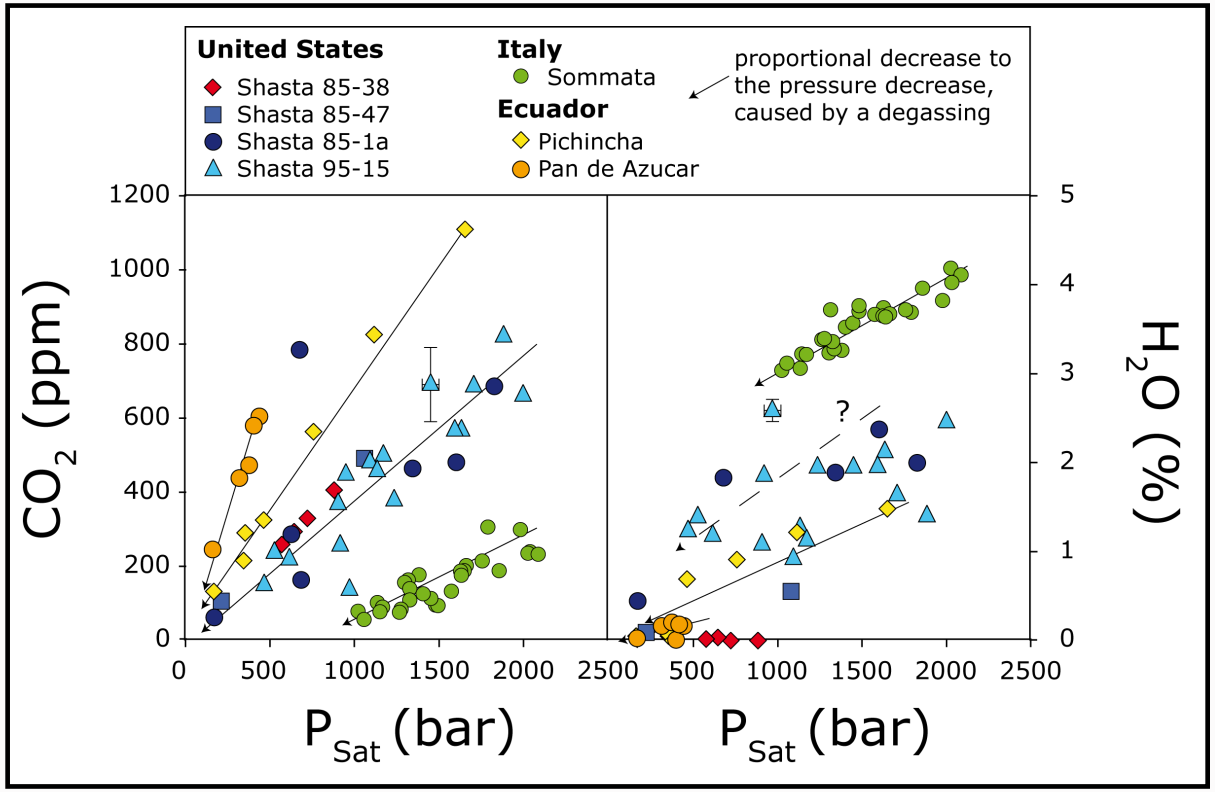
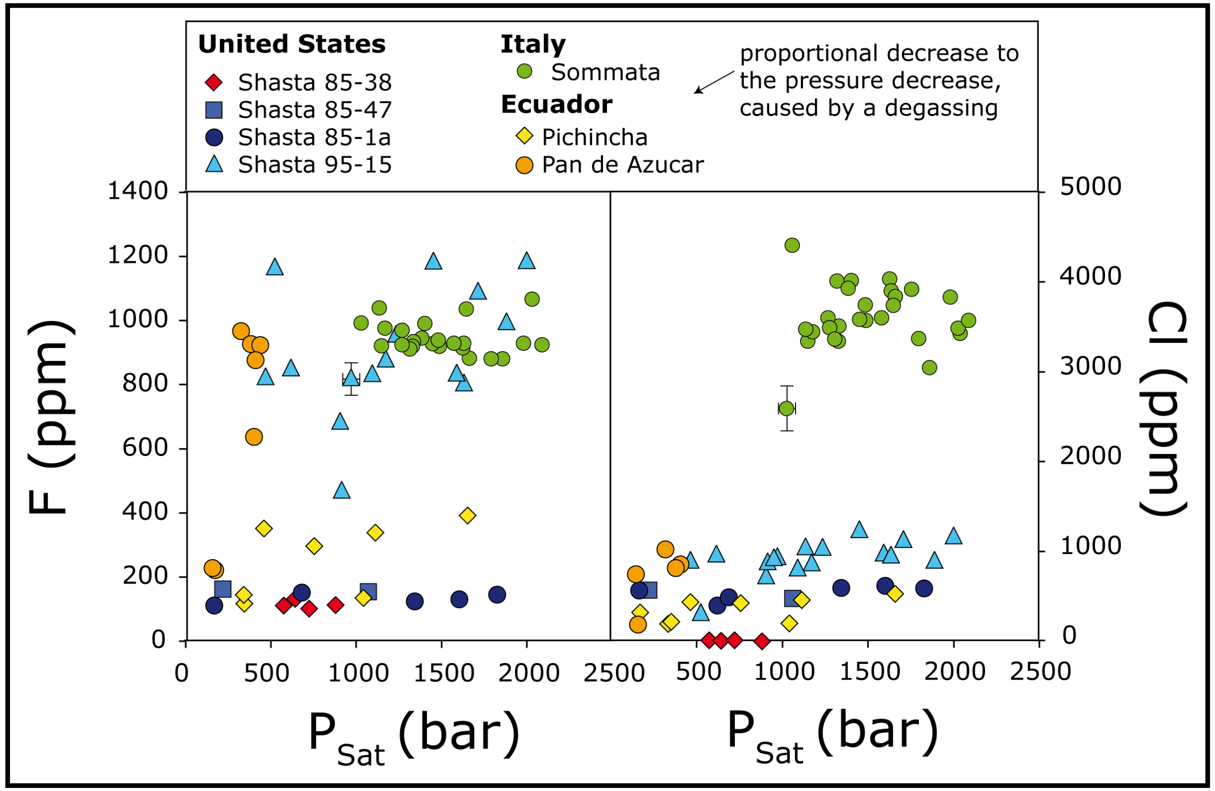




Why F and Cl are way less degassed than H_2O, CO_2 and S ?

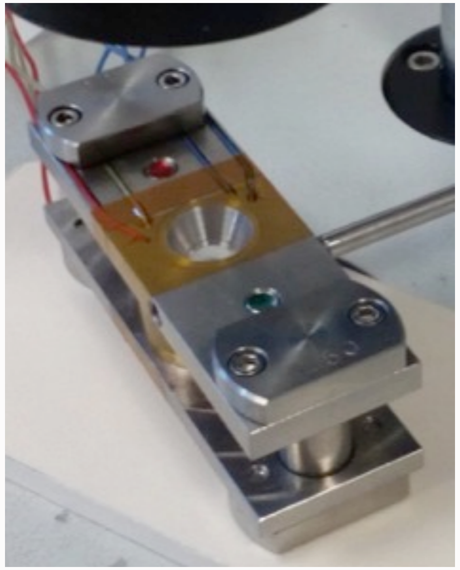
F and Cl degas only during the latest stages of magmatic evolution:
50% of initial Cl in magmas is degassed, compared to only 15% of initial F.

⇒ **WHY ?**

To circumvent barriers raised by examining melts and fluids quenched to ambient *P-T* conditions, *in situ* experimental characterization at *HP-HT* is key.

Hydrothermal diamond anvil cell (HDAC)

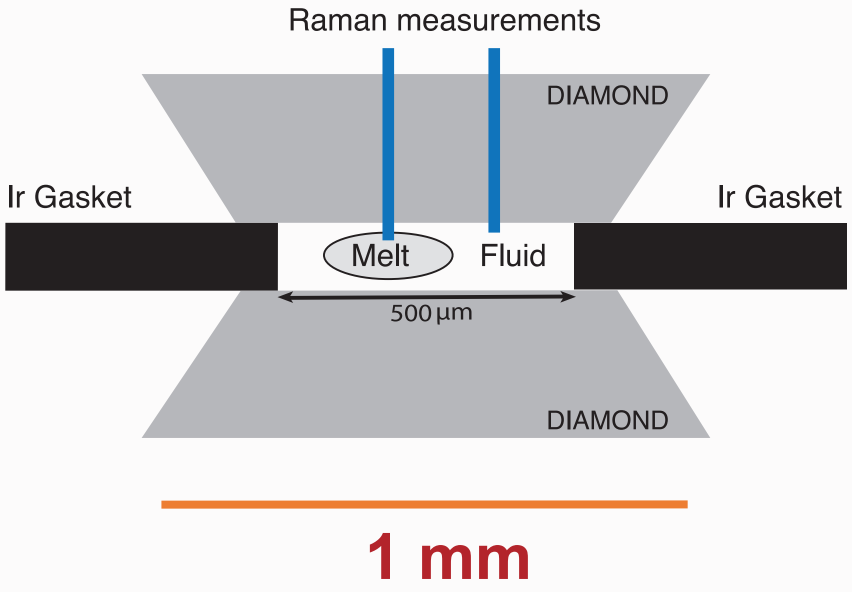
In-situ HP-HT **method to determine volatile element speciation in magmas, minerals, aqueous fluids and gas bubbles (when coupled with Raman spectroscopy)**

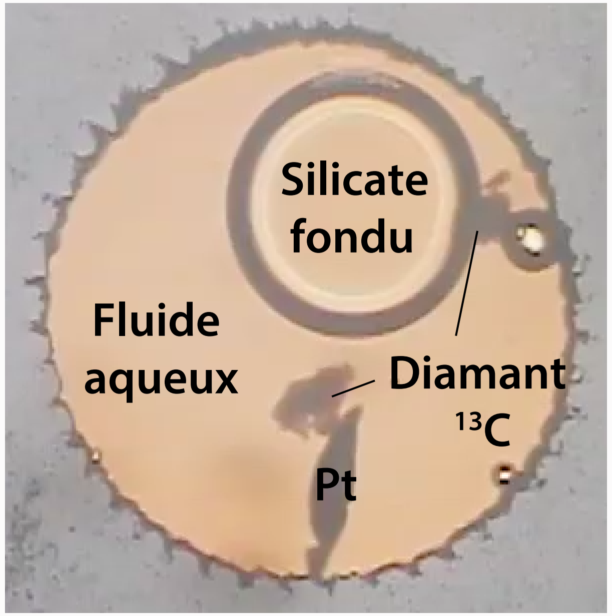


0.3 – 5 GPa

950°C

CRPG





Dalou et al. 2015c

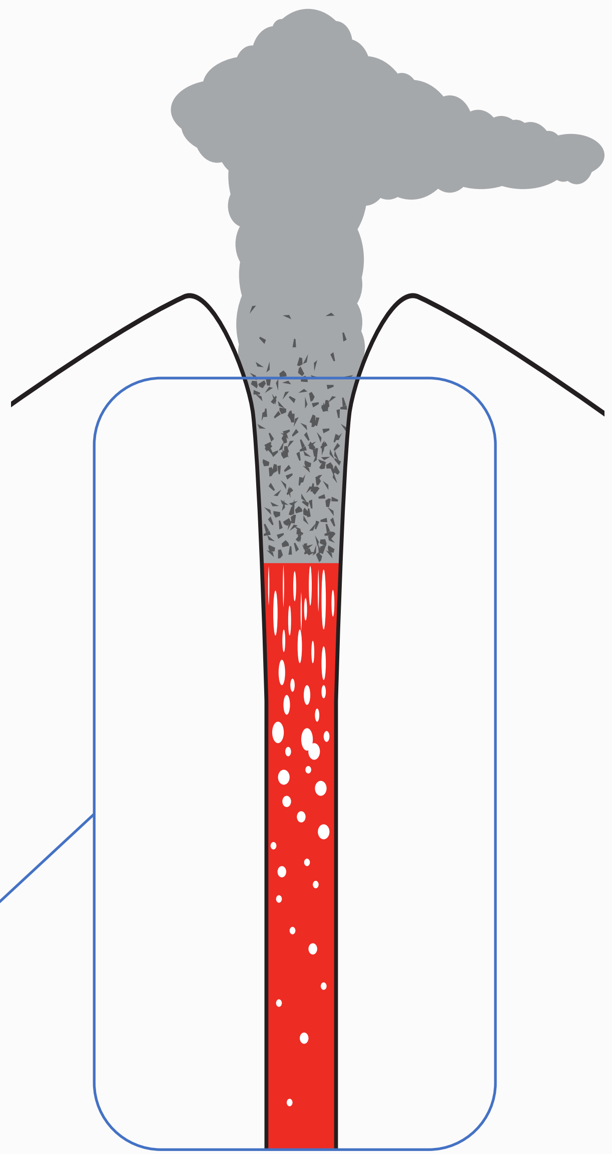
Hydrothermal diamond anvil cell (HDAC)

- Crustal conditions …

- Requires an aqueous fluid phase:
    - Hydrothermalism,
    - Subduction zones

P-T range :

0,1 à 2 GPa – 300 à 950°C



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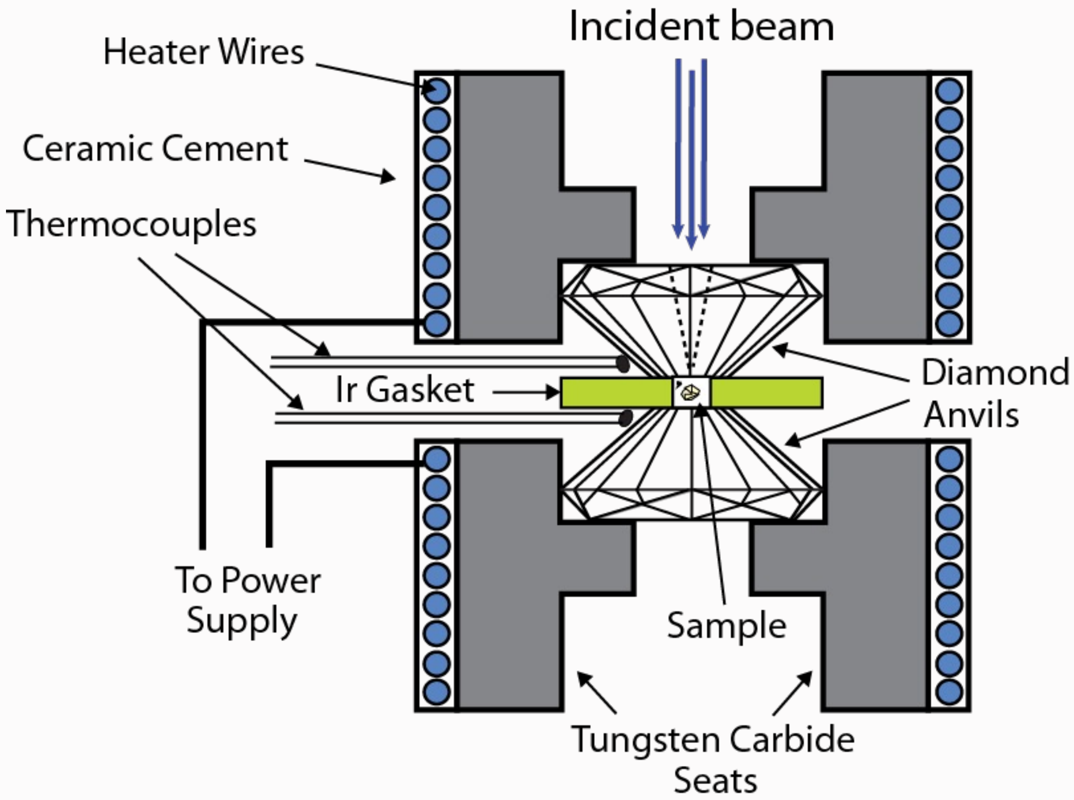

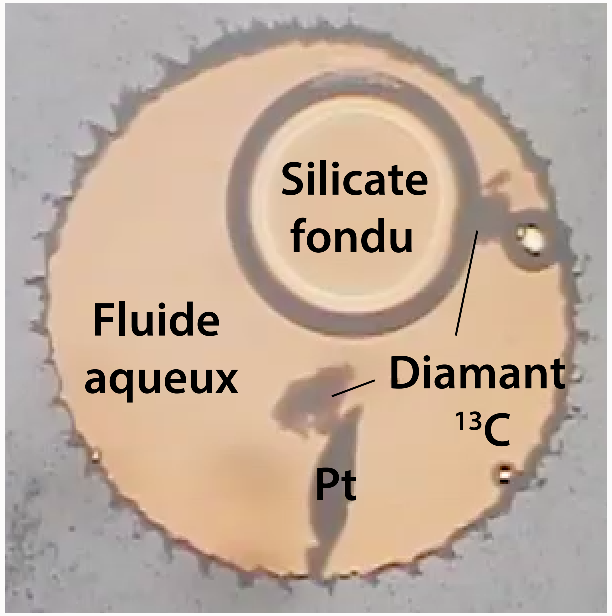

Dalou et al. 2015c

Hydrothermal diamond anvil cell (HDAC)

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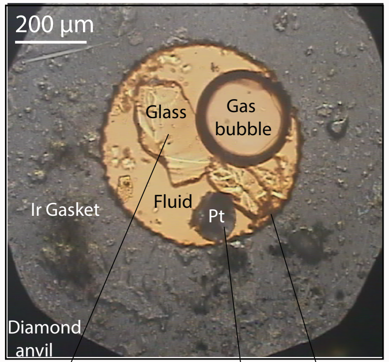
Before the experiment



Glass composition:

$Si_8AlNa_6O_{20}$

NS3A5:
64wt% SiO_2
8.2wt% Al_2O_3
28wt% Na_2O

NBO/T= 0.6

C^{13} diamond to monitor the pressure

rod of Pt metal to enhance D/H equilibration rate

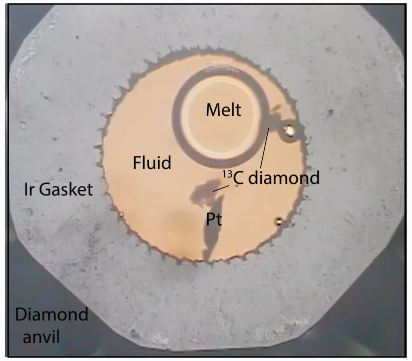
HT

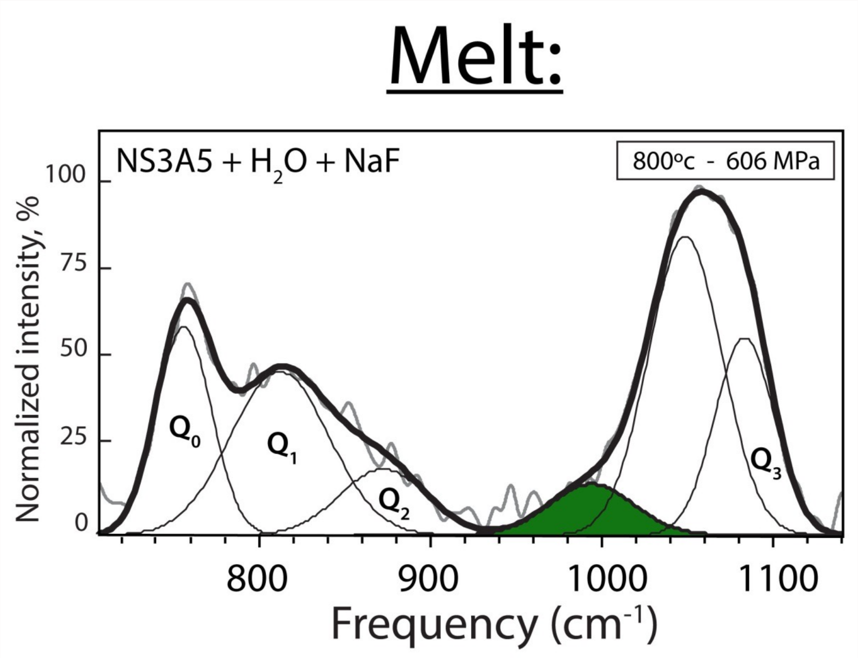
Up to supercritical conditions

One phase: supercritical fluid

HT

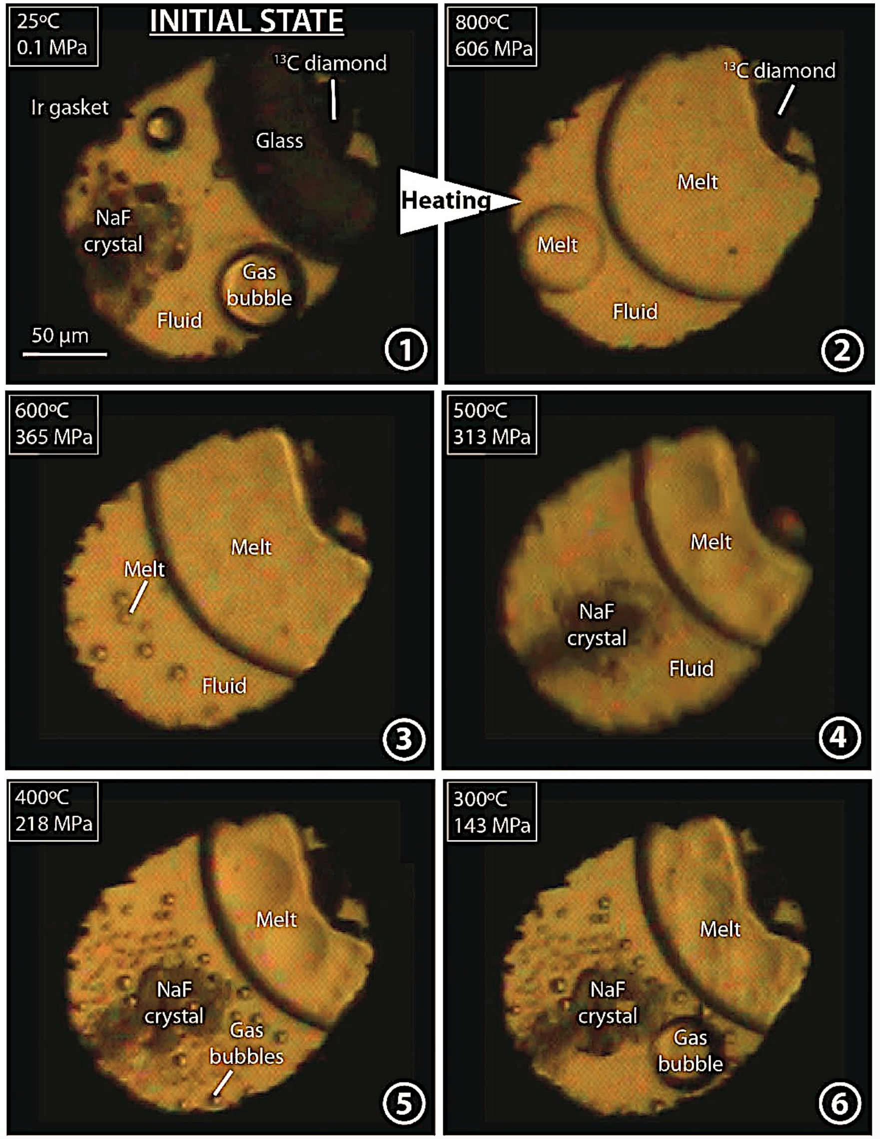
Melt and fluid coexist

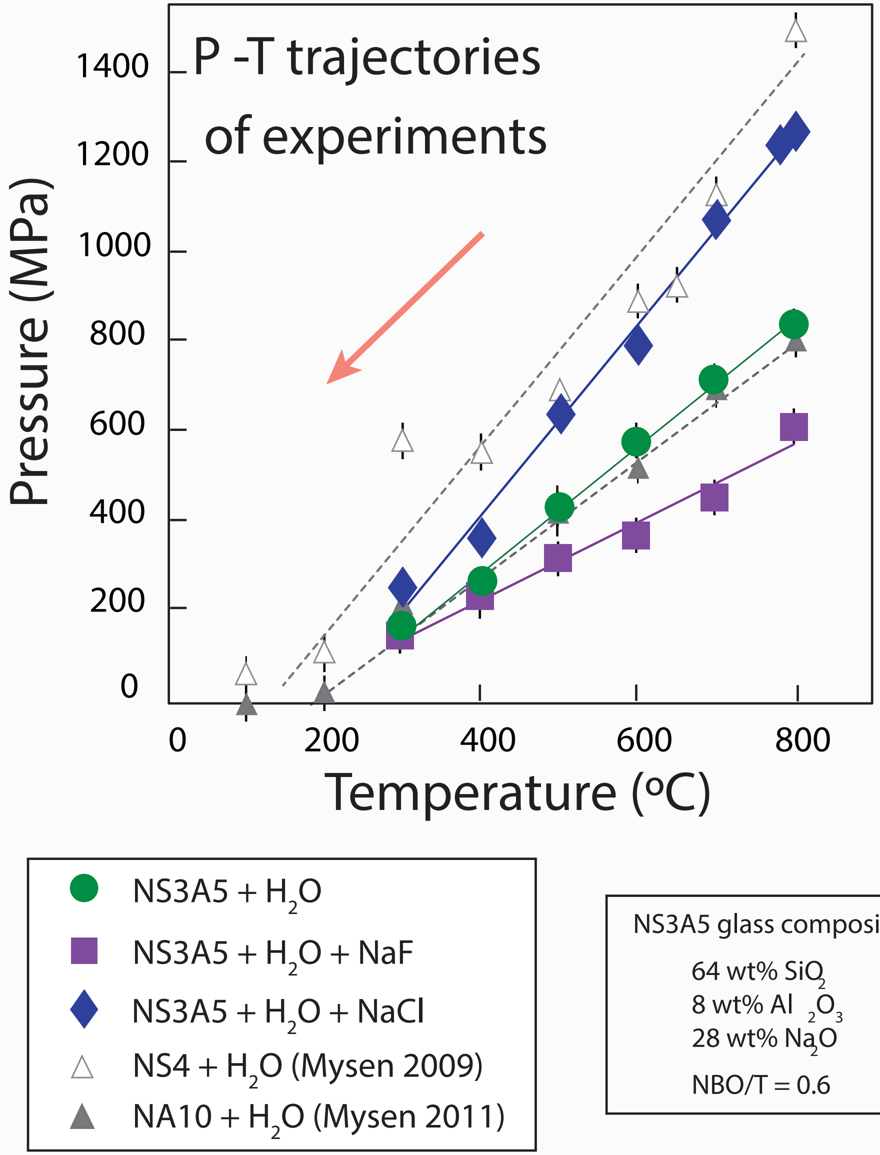




Dalou et al. 2015c

Why F and Cl are way less degassed than H_2O, CO_2 and S ?





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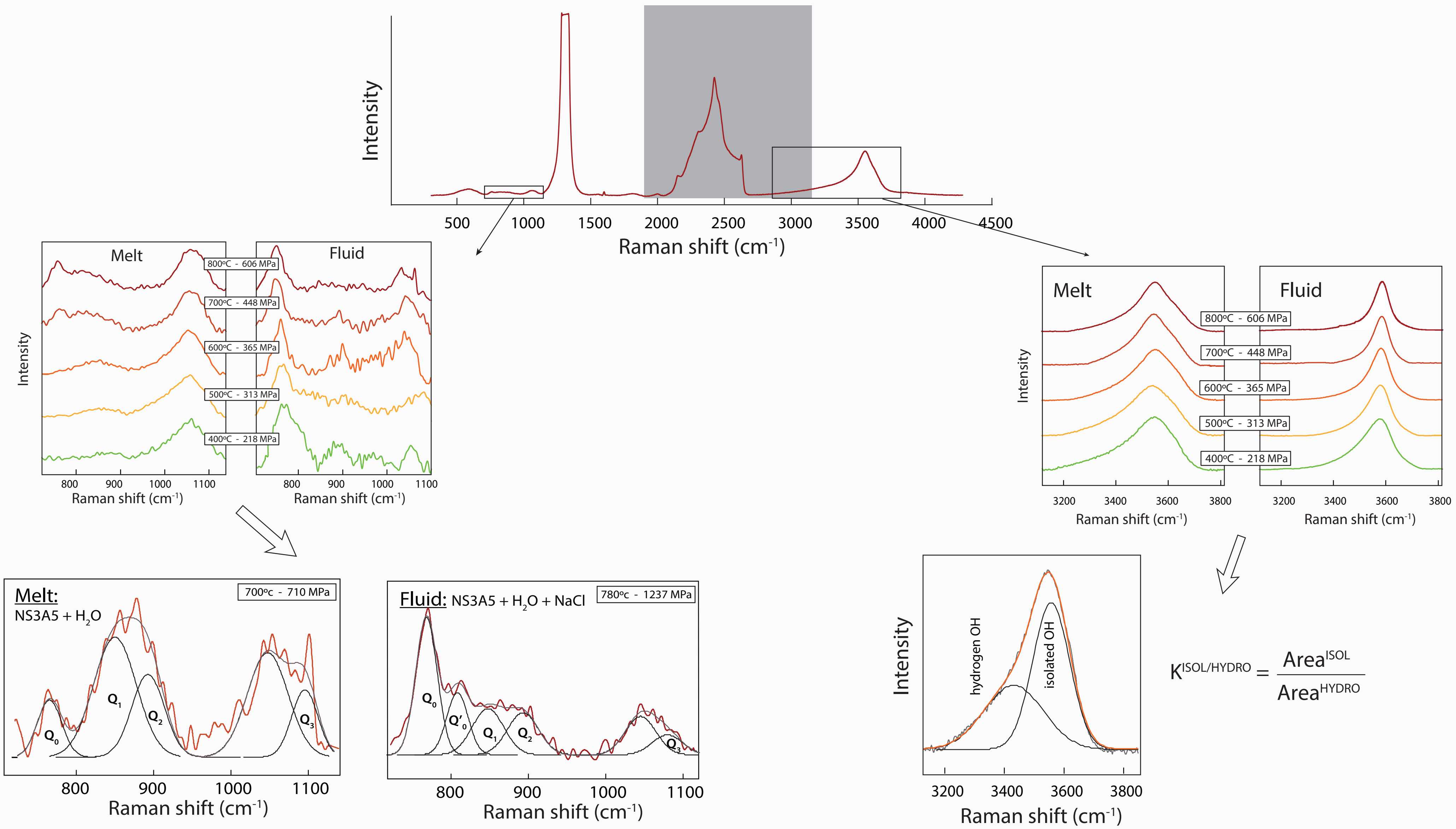

$$K^{ISOL/HYDRO} = \frac{Area^{ISOL}}{Area^{HYDRO}}$$

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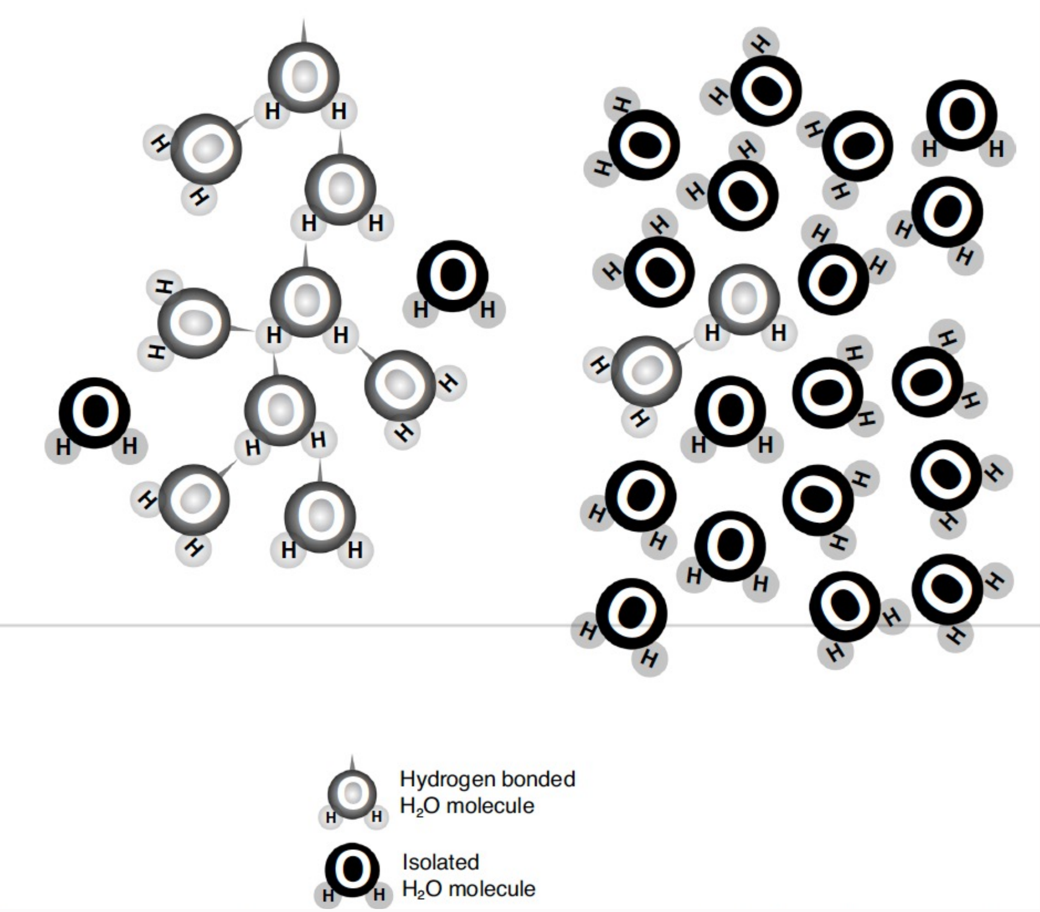
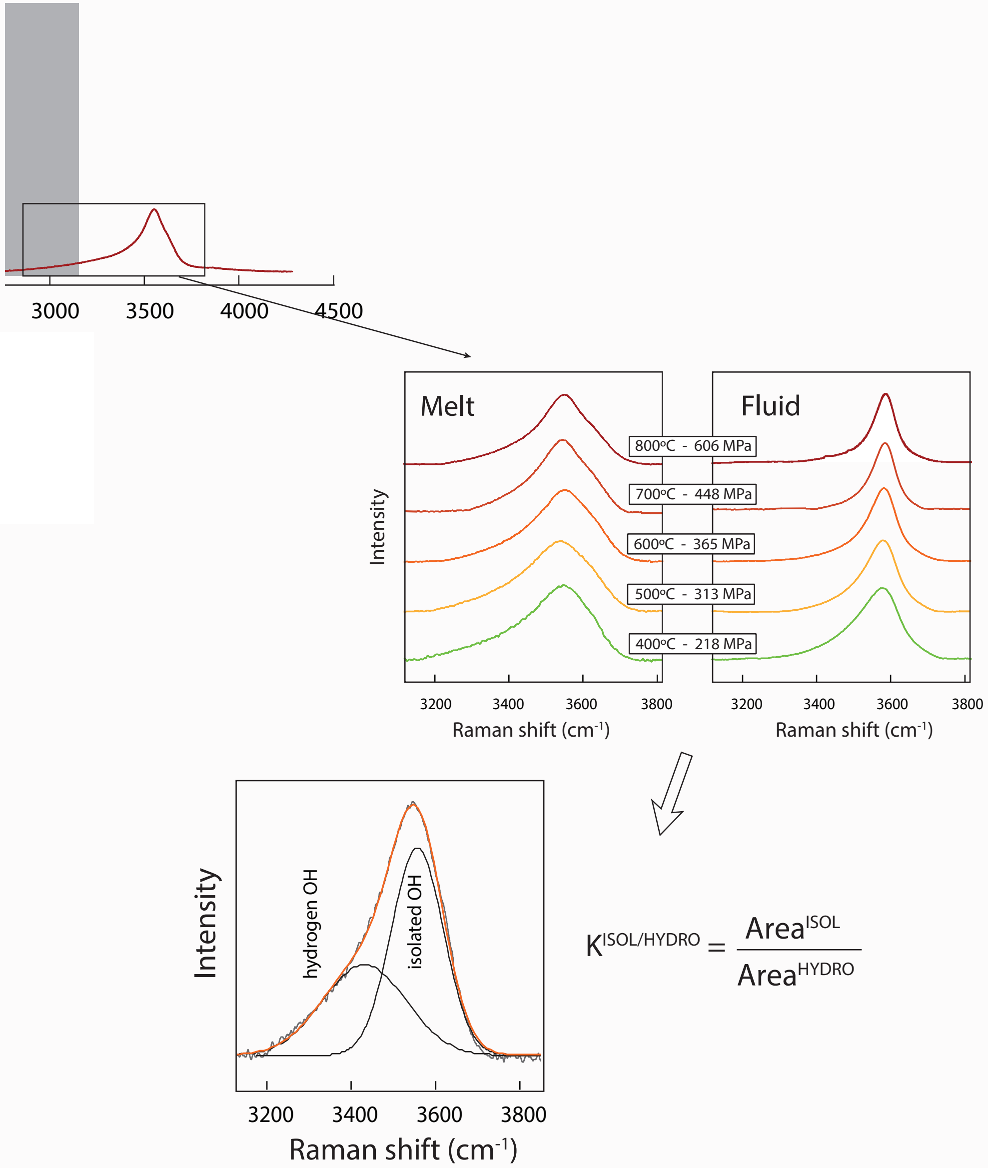


From Mysen (2022)



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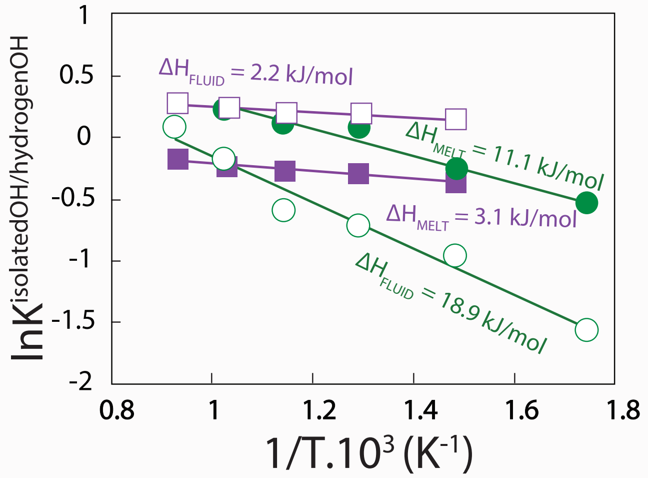
F speciation and fluid/melt partitioning

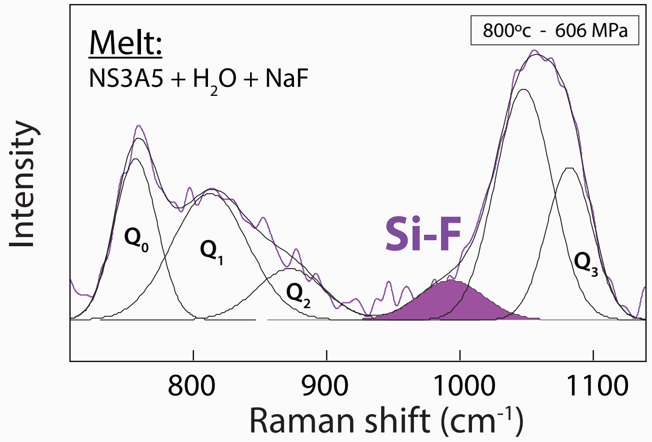


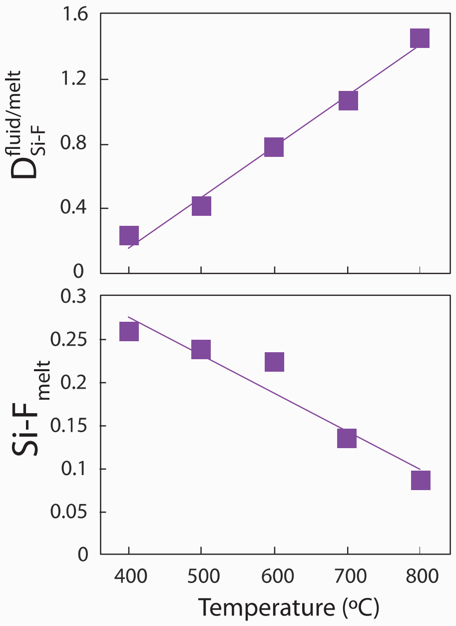
ΔH: enthalpy of rupture of the O···H-O intermolecular bond

$\Delta H_O >> \Delta H_F$

HO-H···F^- bond weaker compared to OH-O···OH_2

$H\text{-}F^-$ bonds are not easy to form in fluids or melts



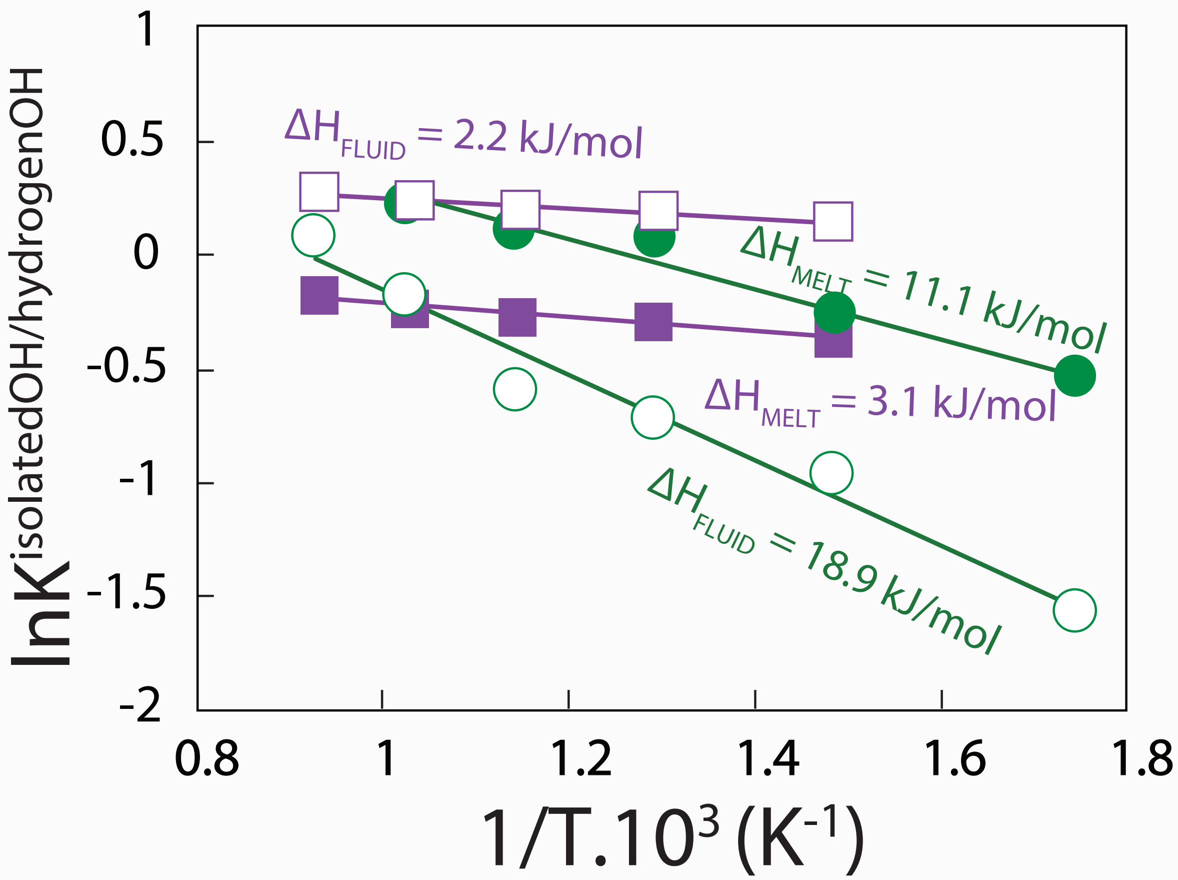


Increasing $D^{fluid/melt}_{Si\text{-}F}$ with increasing T

Si-F complexes increasingly partition from the fluid into the melt during cooling and ascension

Why F and Cl are way less degassed than H_2O, CO_2 and S ?

F speciation and fluid/melt partitioning



➡

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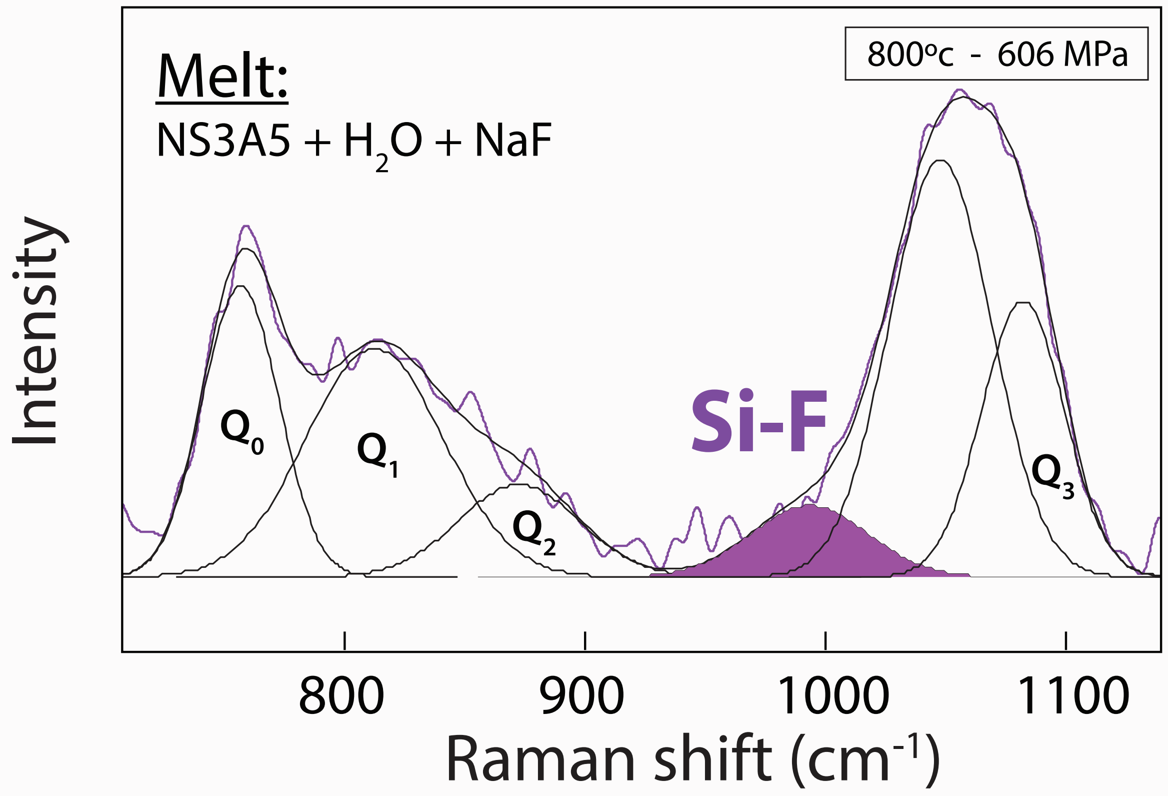

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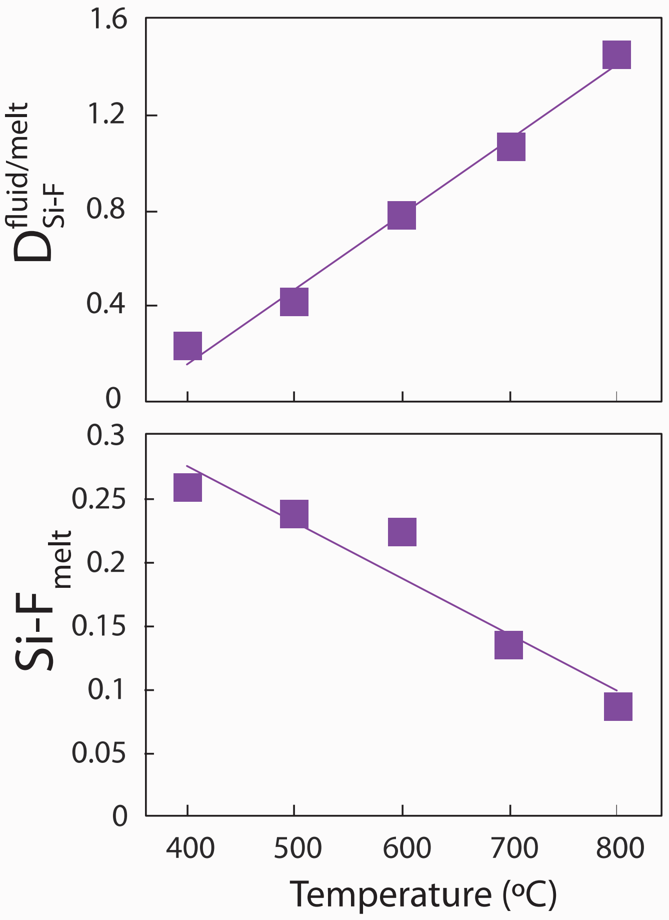

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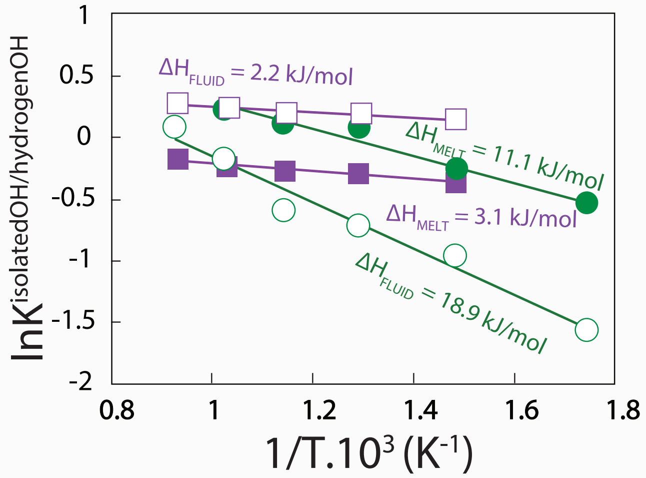

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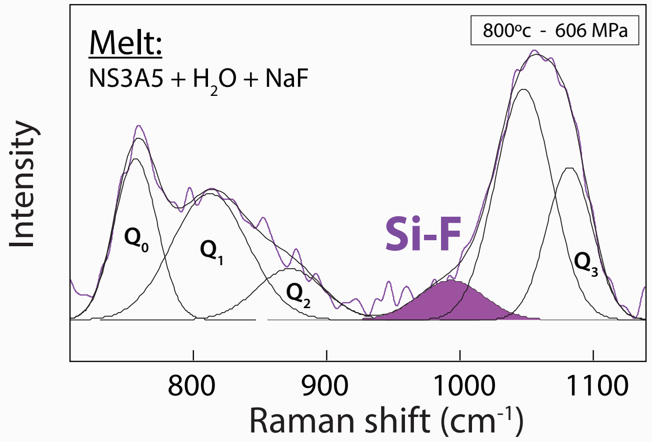

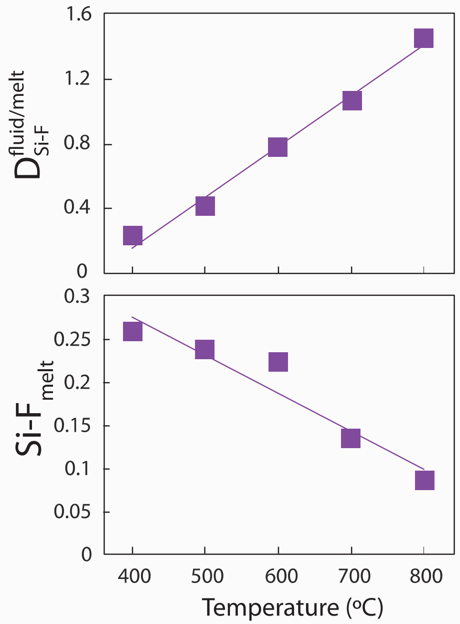

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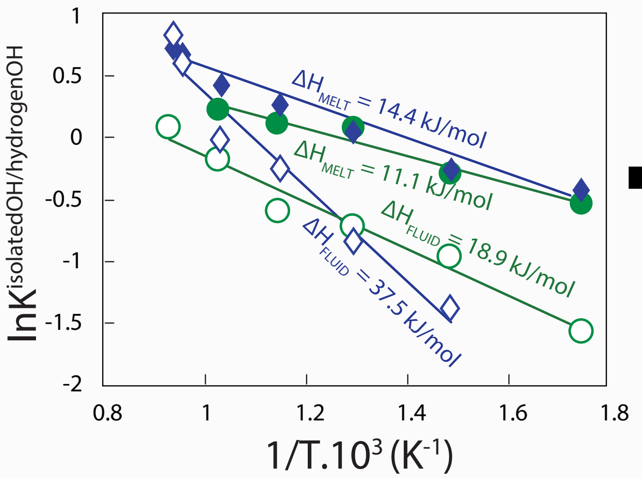
Cl speciation and fluid/melt partitioning



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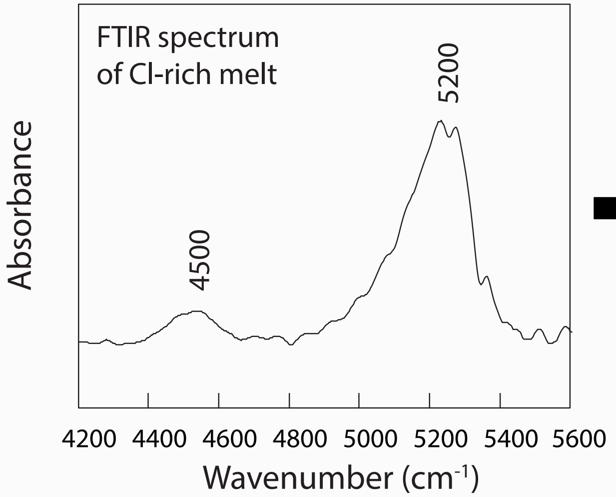
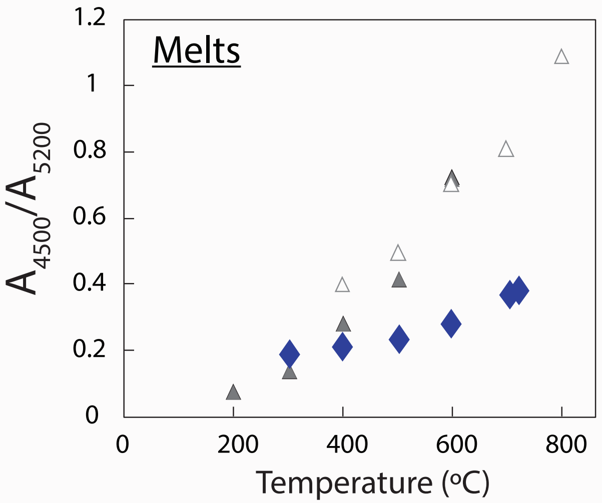
HO-H· · ·Cl⁻ bond stronger than OH-O· · ·OH_2 bond
(Cl⁻· · ·H electrostatic attraction
is stronger than O· · ·H bonds)

H-Cl⁻ bonds are easy to form, particularly in fluids





Cl-bearing melts show lower OH/H_2O ratiosthan halogen-free melts

⇒ explained by the equilibrium reaction:

$$2\,NaOH + Q^4 + Cl_2 \Leftrightarrow 2\,NaCl + Q^2 + H_2O$$

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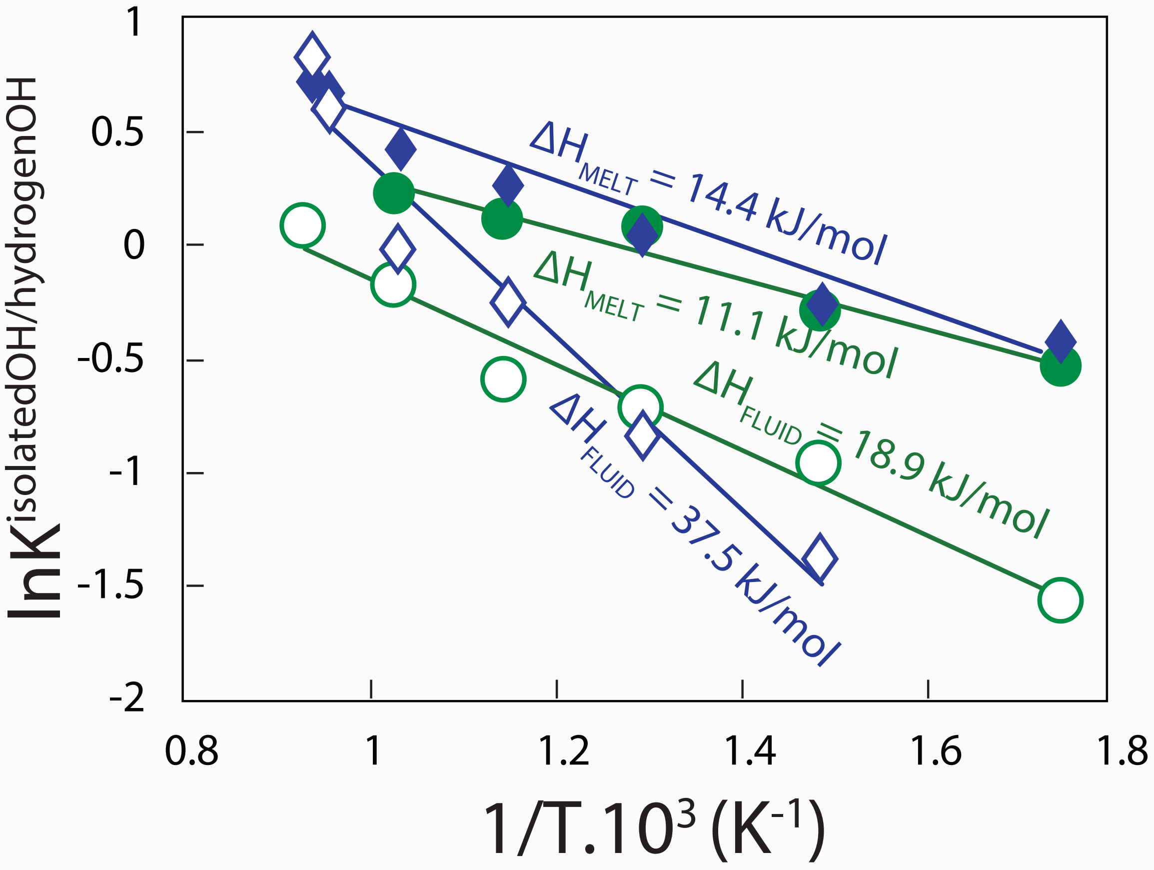

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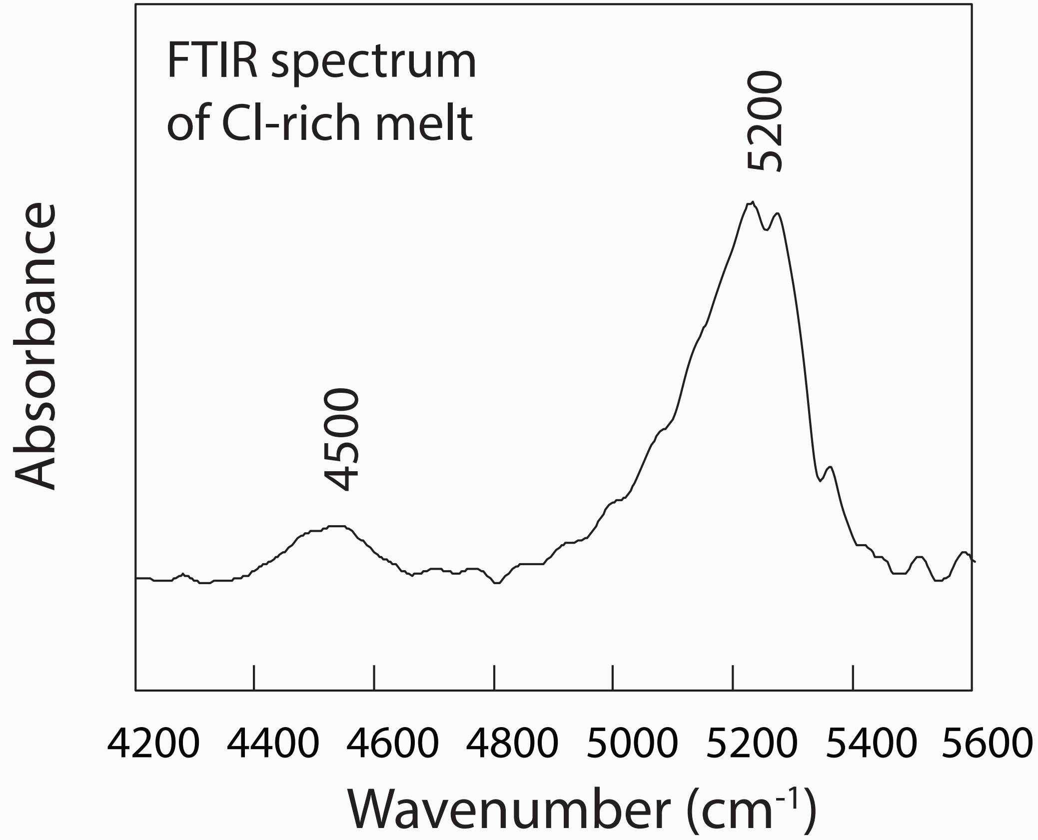

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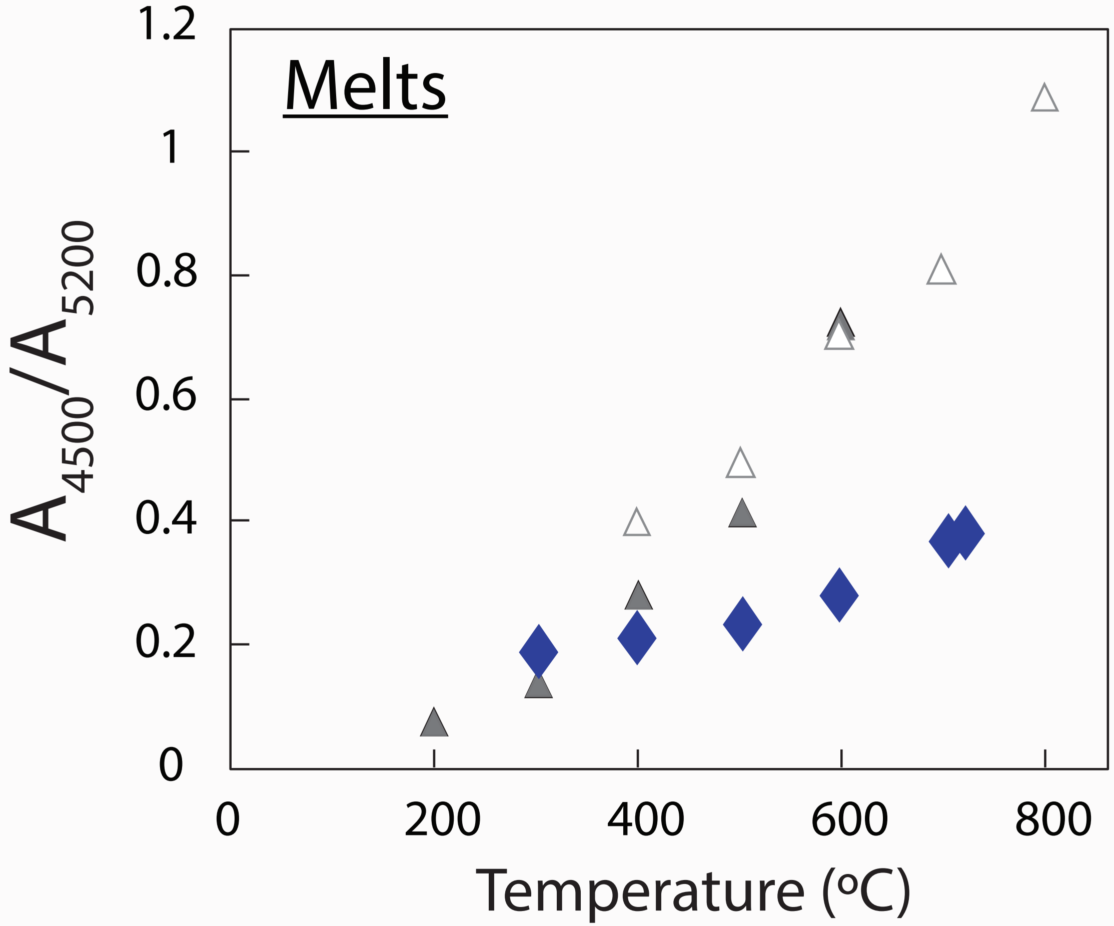

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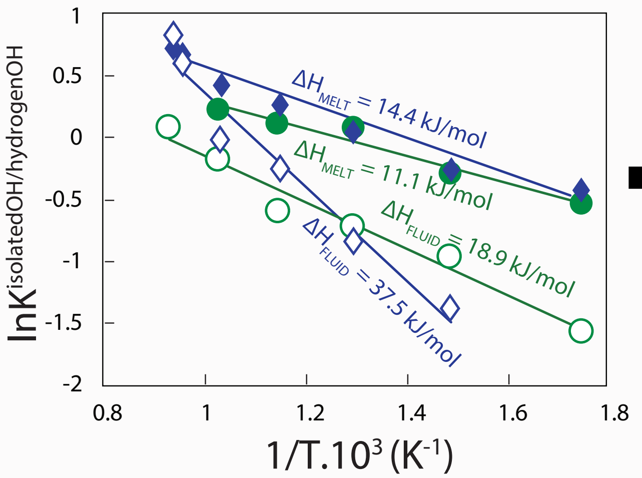

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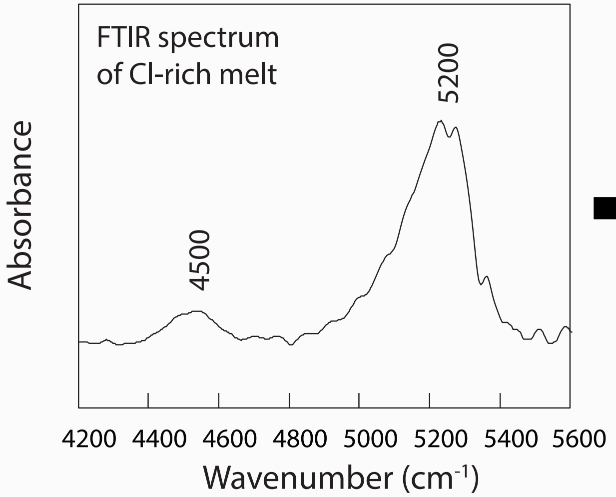

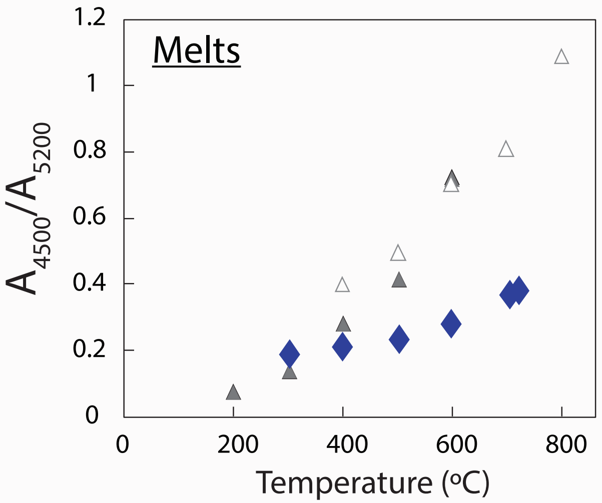

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Why F and Cl are way less degassed than H_2O, CO_2 and S ?

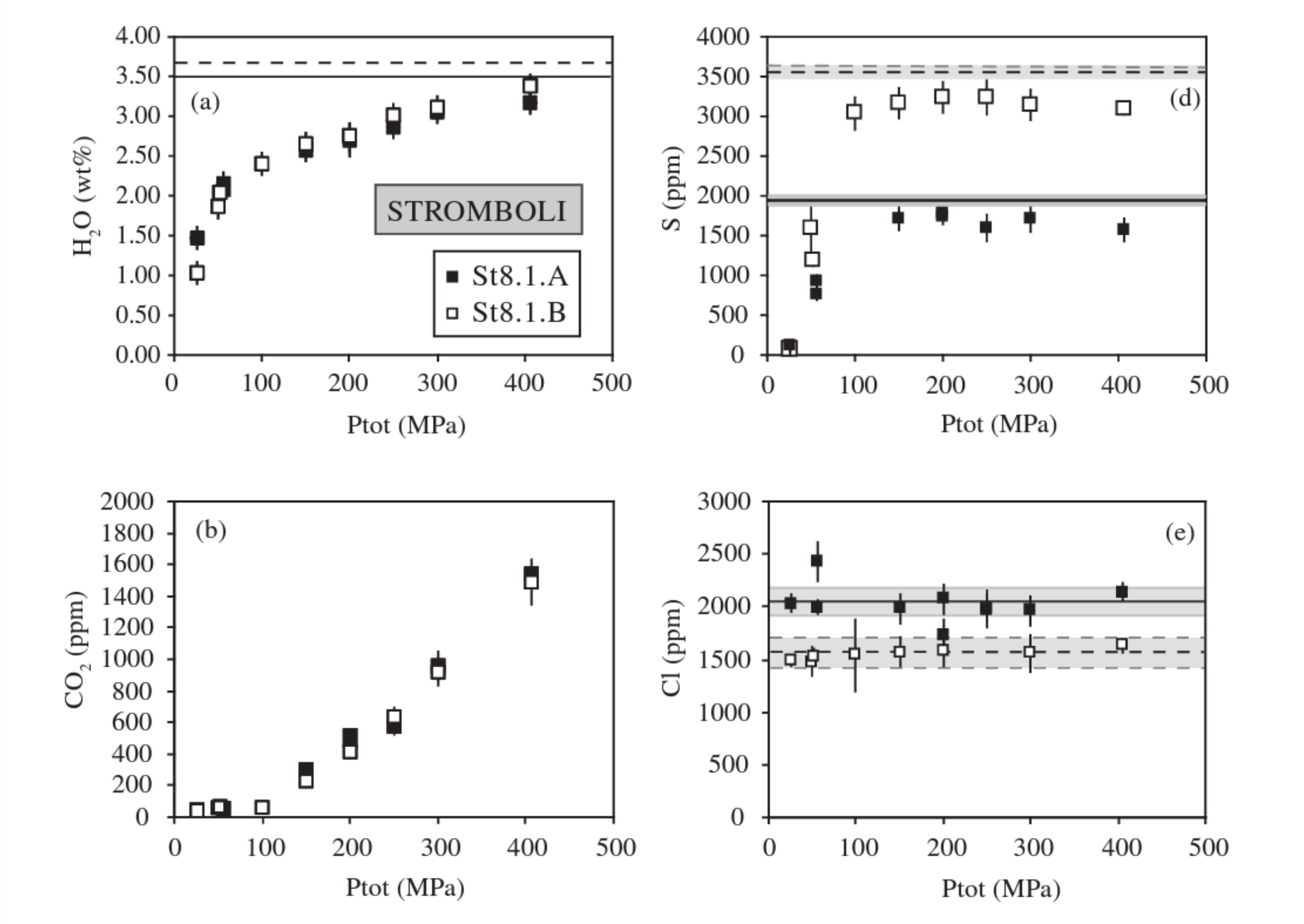
- F solubility remains high in magmas during their ascent because of the Si-F bonds it forms.

- In constrast, Cl forms Na-Cl bonds in magmas and magmatic fluids, but HCl may be the dominant species in fluids above 790 MPa.

- F solubility in magmas remains higher than Cl likely due to the increasing formation of HCl in the last ~5–25 km of decompression.

Why F and Cl are way less degassed than H_2O, CO_2 and S ?

- In comparison :
- H2O
- CO2
- S



Lesne et al. (2011)

Take home message

F and Cl bonding in the silicate network → F and Cl soluble in magmas up to shallow depth

Poorly or no degassed in melt inclusions

F and Cl contents in MI → primary magma

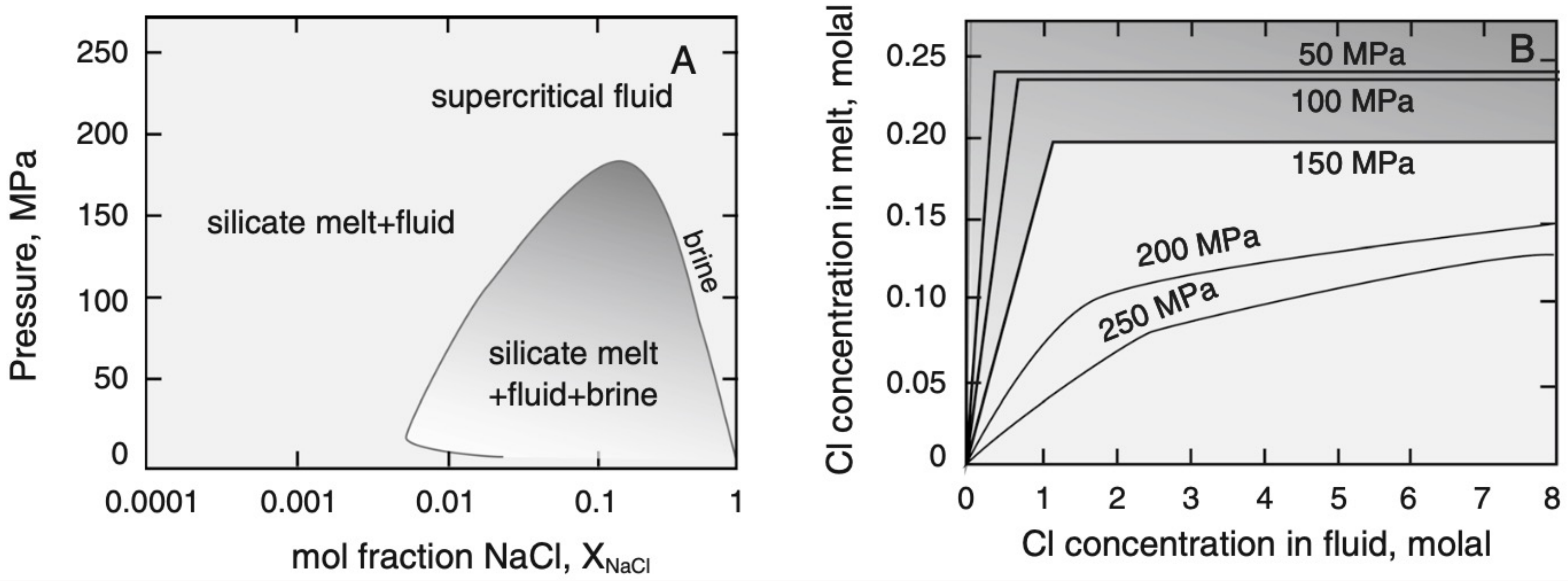
F/Cl ratios in arc melt inclusions are highly variable

D_F and D_{Cl} sensitive to water content in the melt

May reflect various hydrous flux processes in the mantle wedge

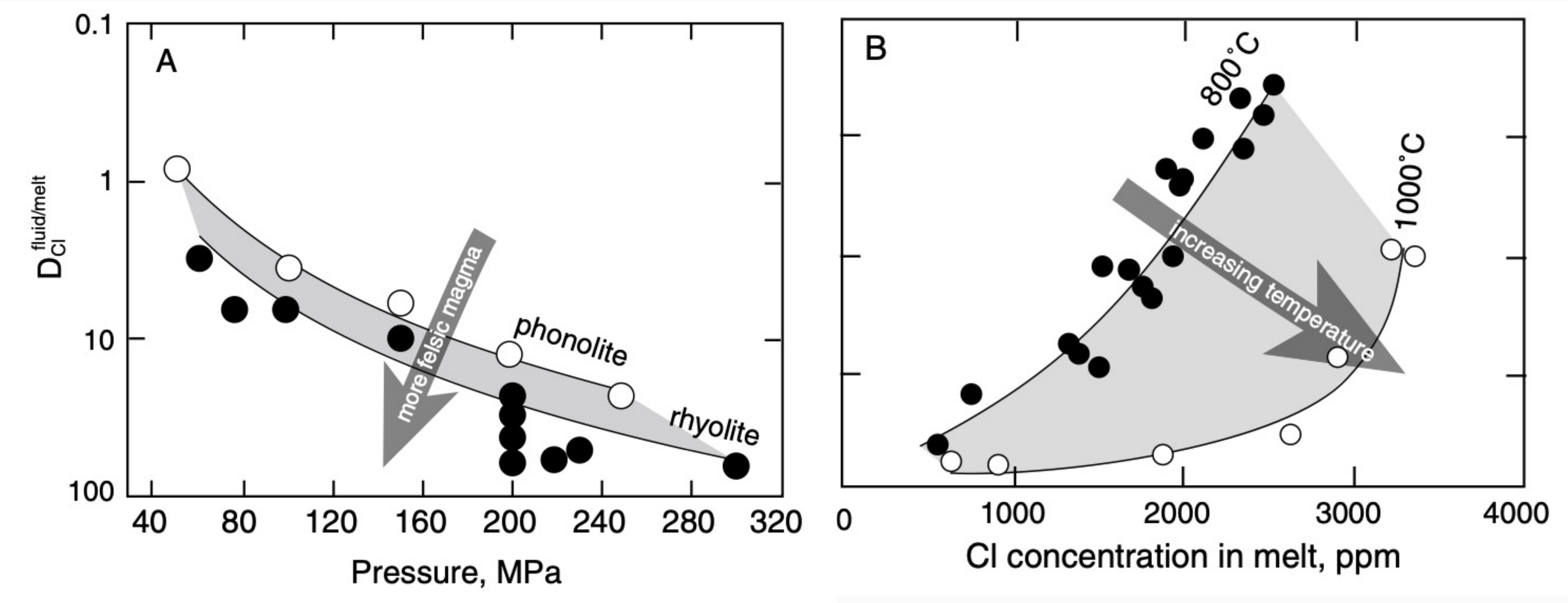
THANKS FOR YOUR ATTENTION

Back up slides



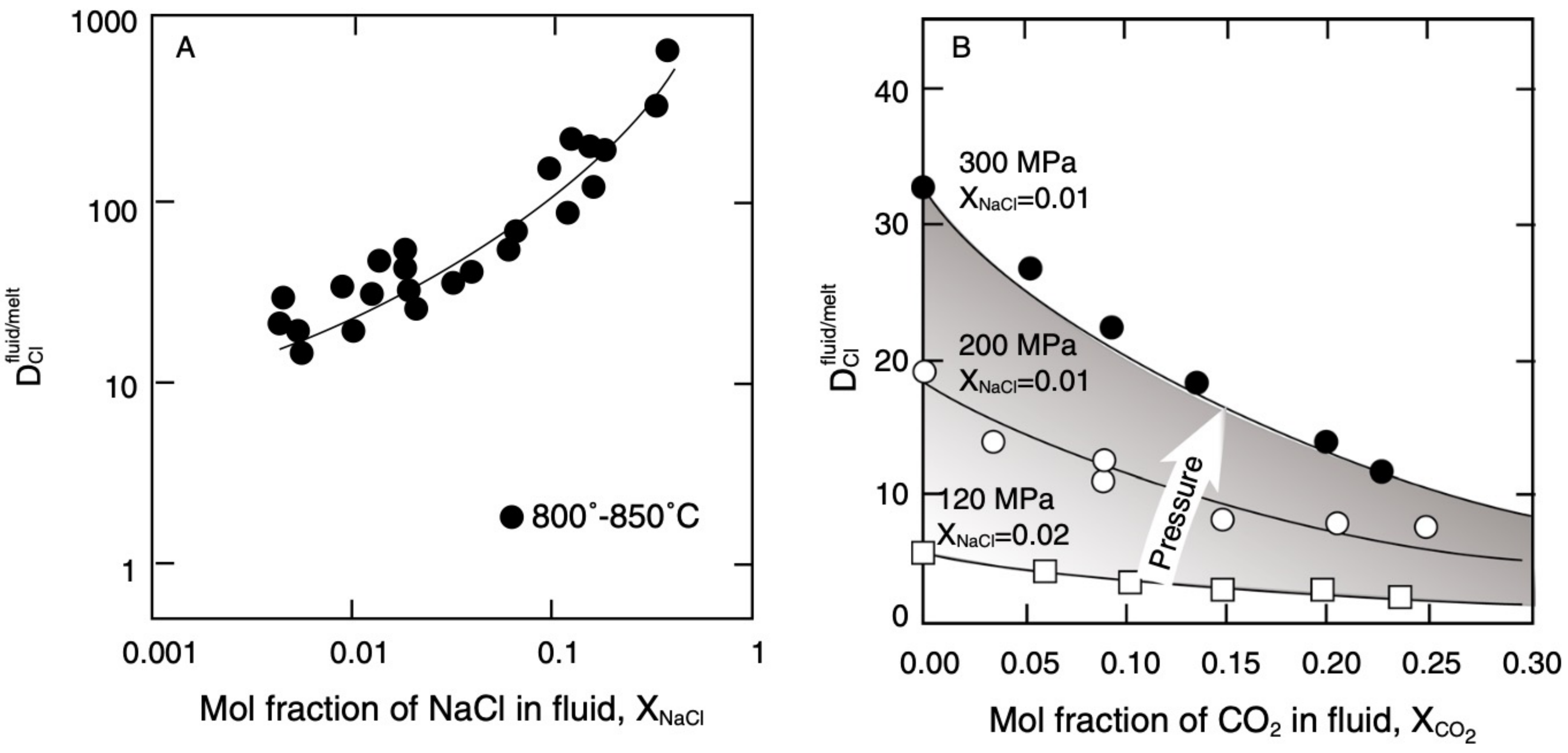
A
supercritical fluid
silicate melt+fluid
brine
silicate melt
+fluid+brine
Pressure, MPa
250
200
150
100
50
0
0.0001
0.001
0.01
0.1
1
mol fraction NaCl, X_{NaCl}
B
50 MPa
100 MPa
150 MPa
200 MPa
250 MPa
Cl concentration in melt, molal
0.25
0.20
0.15
0.10
0.05
0
0
1
2
3
4
5
6
7
8
Cl concentration in fluid, molal

Mysen (2022)

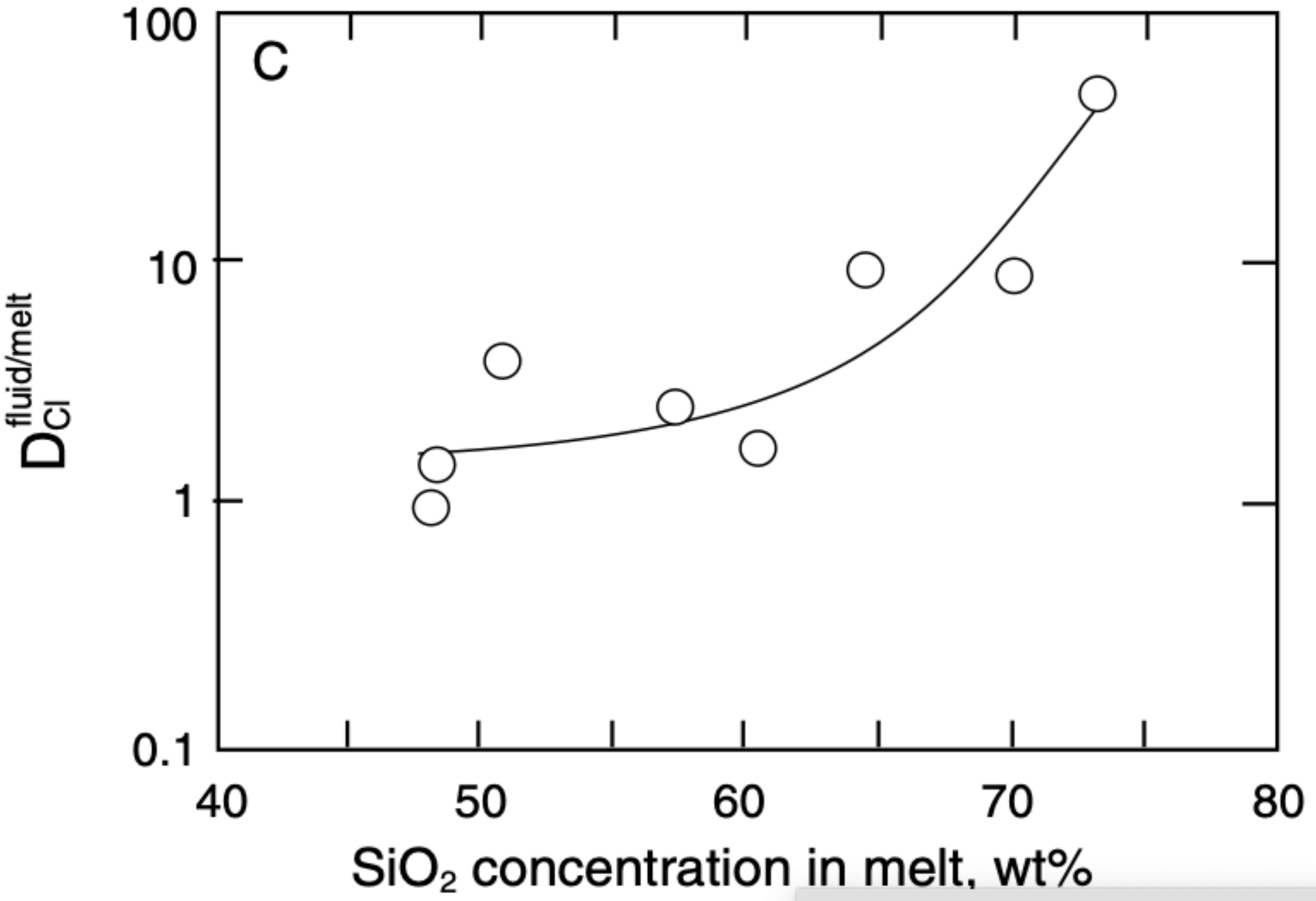


0.1
1
10
100
A
$D_{Cl}^{fluid/melt}$
more felsic magma
phonolite
rhyolite
40
80
120
160
200
240
280
320
Pressure, MPa
B
800°C
1000°C
increasing temperature
0
1000
2000
3000
4000
Cl concentration in melt, ppm

Mysen (2022)

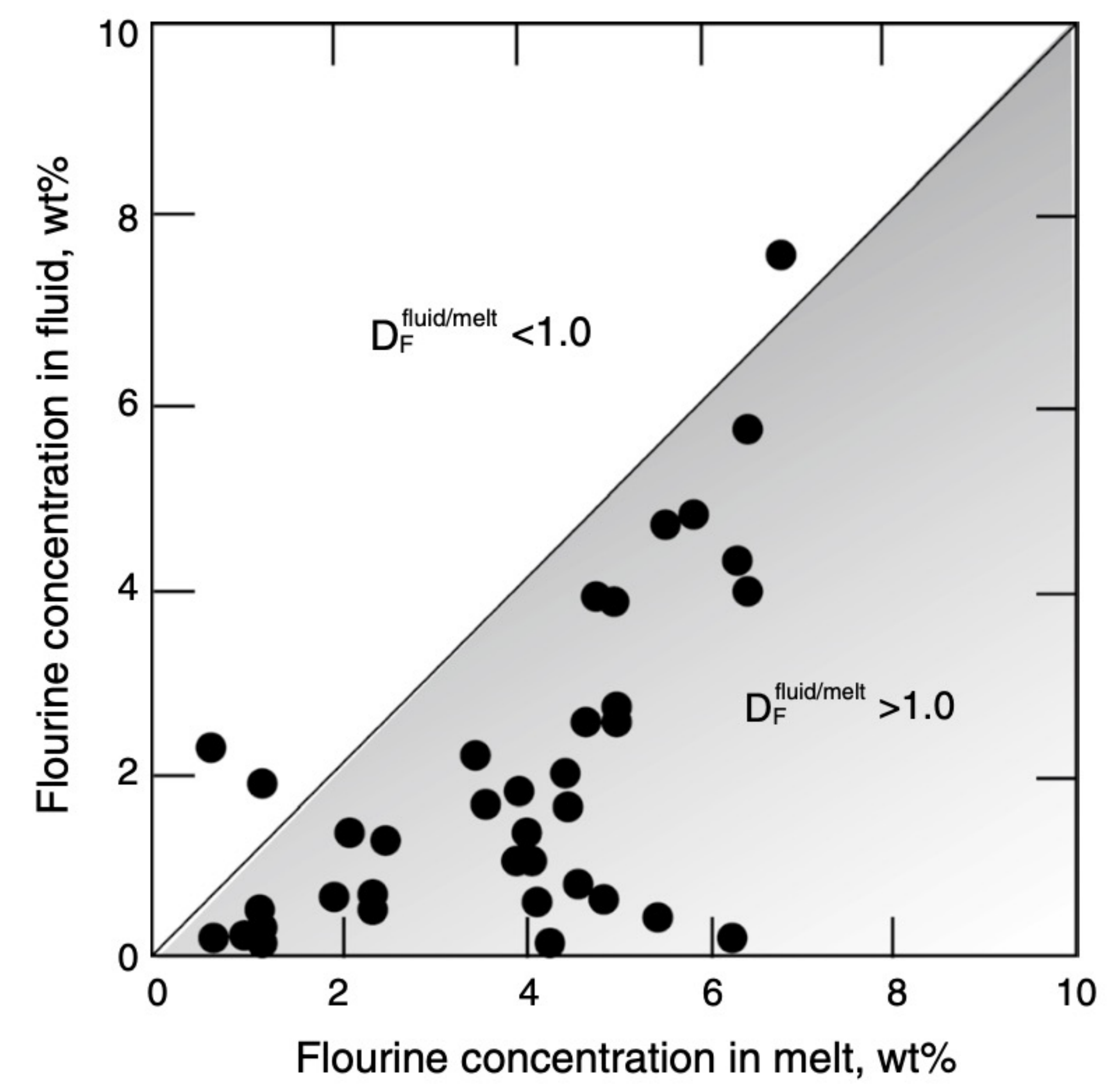

A
1000
100
10
1
$D_{Cl}^{fluid/melt}$
800˚-850˚C
0.001
0.01
0.1
1
Mol fraction of NaCl in fluid, X_{NaCl}
B
40
30
20
10
0
300 MPa
X_{NaCl}=0.01
200 MPa
X_{NaCl}=0.01
120 MPa
X_{NaCl}=0.02
Pressure
0.00
0.05
0.10
0.15
0.20
0.25
0.30
Mol fraction of CO_2 in fluid, X_{CO_2}

Mysen (2022)



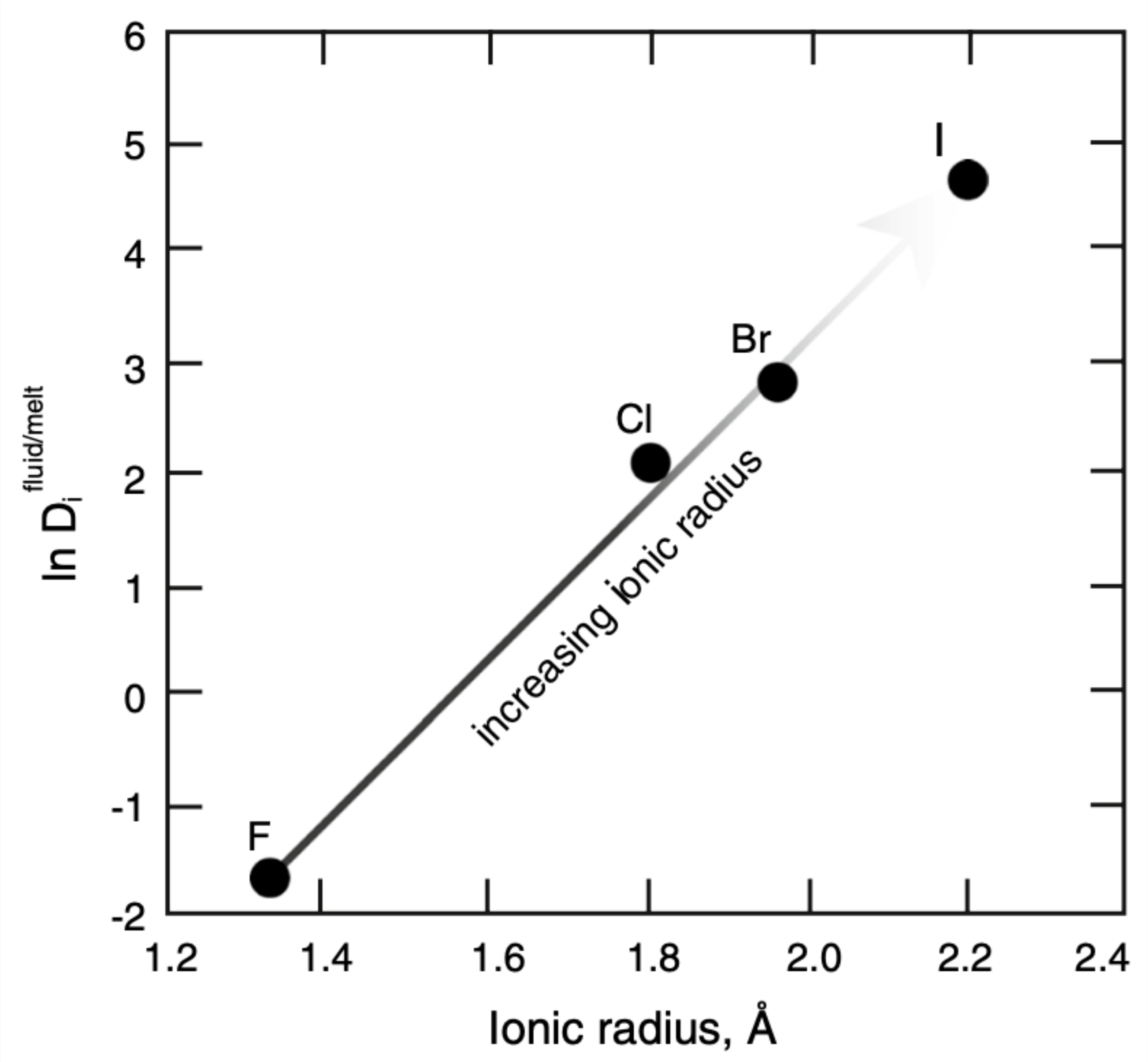
100
10
1
0.1
C
$D_{Cl}^{fluid/melt}$
40
50
60
70
80
SiO_2 concentration in melt, wt%

Mysen (2022)



10
8
6
4
2
0
0
2
4
6
8
10
$D_F^{fluid/melt}$ <1.0
$D_F^{fluid/melt}$ >1.0
Flourine concentration in fluid, wt%
Flourine concentration in melt, wt%

Mysen (2022)



ln $D_i^{fluid/melt}$
Ionic radius, Å
F
Cl
Br
I
increasing ionic radius
6
5
4
3
2
1
0
-1
-2
1.2
1.4
1.6
1.8
2.0
2.2
2.4

Mysen (2022)