

# MINERAL CHEMISTRY OF THE *LLAIMA* PUMICE, SOUTHERN CHILE: EVIDENCE FOR MAGMA MIXING

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(4 figures, 1 plate, 1 table)

**ABSTRACT.** This paper details the mineralogy and the chemistry of the *Llaima* pumice deposit from in southern Chile. This andesitic to dacitic pumice was erupted from *Llaima* Volcano. This eruption took place in the early Holocene through at least two pulses that are well recorded in deposits around the Icalma-Galletué area. In 2001, the *Llaima* pumice was sampled from six outcrops. The mineralogy has been studied under polarizing microscope and electron microprobe (EPMA). The *Llaima* pumice contains phenocrystals of orthopyroxene, clinopyroxene, olivine and titanomagnetite. The olivine is characterized by two chemically distinguishable populations (*i.e.* Fo<sub>32</sub> to Fo<sub>35</sub> and Fo<sub>76</sub> to Fo<sub>82</sub>). The respective chemical compositions of these two distinct olivines emphasize that the eruption was triggered by magma mixing.

**Keywords:** Chile, Lake District, Holocene, volcano, pumice, olivine

**RÉSUMÉ. Chimie des minéraux de la ponce *Llaima*, Sud du Chili : Evidence d'un mélange magmatique.** Ce travail détaille les compositions minéralogique et chimique de la ponce *Llaima*. Celle-ci est andésitique à dacitique, et résulte d'une éruption du volcan *Llaima* (Naranjo & Moreno, 1991). Cette éruption s'est effectuée en deux temps durant la première moitié de l'Holocène. En 2001, la ponce *Llaima* a été échantillonnée dans six coupes. Elle est étudiée au microscope polarisant et à la microsonde électronique. La ponce *Llaima* contient des phénocristaux d'orthopyroxène, de clinopyroxène, d'olivine et de titanomagnétite. Les deux populations d'olivine, chimiquement distinguables (*i.e.* Fo<sub>32</sub> à Fo<sub>35</sub> et Fo<sub>76</sub> à Fo<sub>82</sub>) attestent que l'éruption fut déclenchée par un mélange de magma.

**Mots-clés :** Chili, Région des Lacs, Holocène, volcan, ponce, olivine

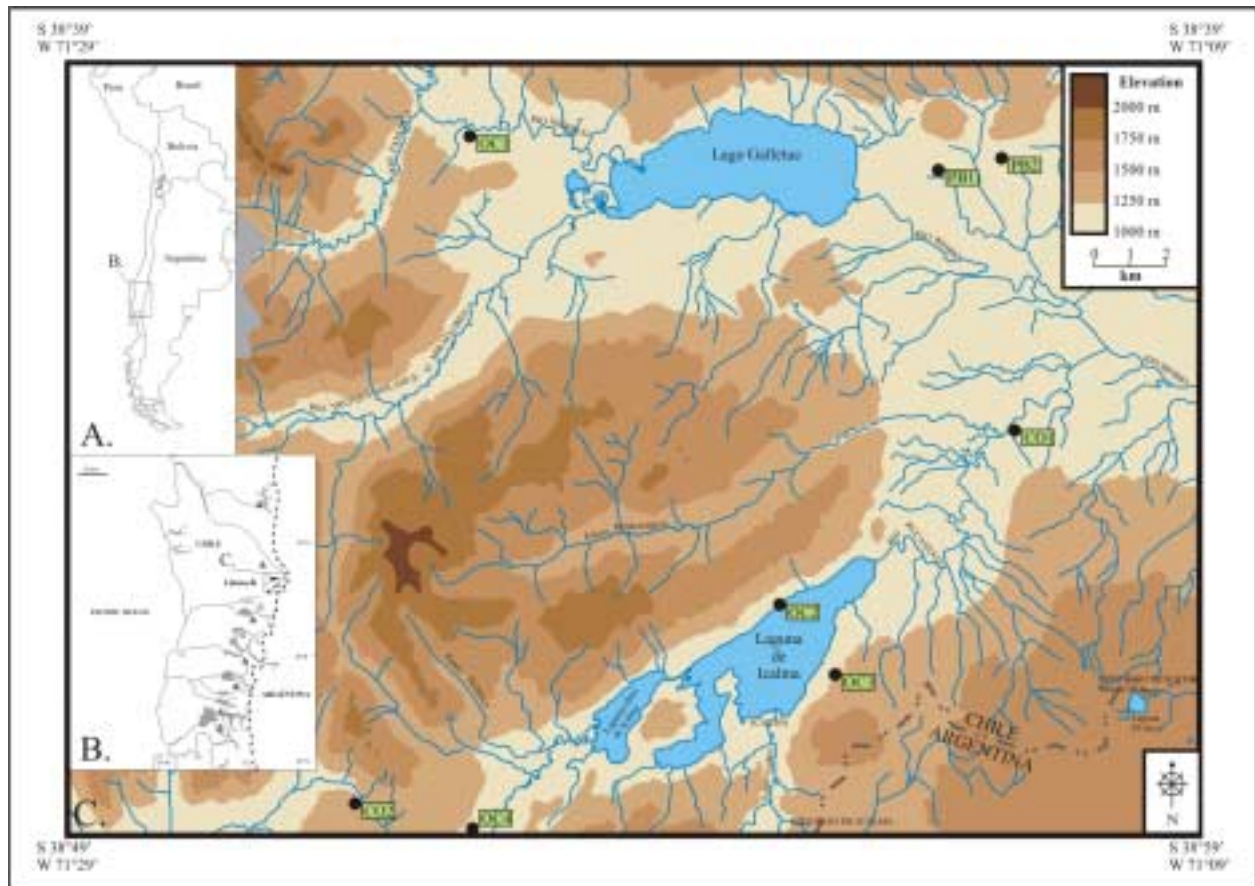
## 1. Introduction

The studied area is located in the central part of the Southern Volcanic Zone (SVZ) (López-Escóbar *et al.*, 1977; Thorpe & Francis, 1979; López-Escóbar, 1984). The SVZ rocks are dominated by basalts with subsidiary basaltic andesites and dacites rich in orthopyroxene, clinopyroxene and Fe-rich olivine (López-Escóbar, *op. cit.*; Futa & Stern, 1988). The occurrence of Fe-rich olivine is sometimes interpreted as resulting from partially assimilated inclusions of basaltic material (Gerlach *et al.*, 1988).

Lakes Icalma and Galletué are two glacial lakes (~4 km long) located in *la Región de la Araucanía* (Chile, 37°0' – 39°30' S; Fig. 1). They are surrounded westwards by a volcanic field that has been active throughout the Quaternary (*e.g.* Naranjo & Moreno, 1991; Naranjo *et al.*, 1993; Gonzalez-Ferran, 1995). Hence a lot of tephra falls were transported by dominating *Westerlies* winds. The eruptions that emplaced these tephra falls are well recorded in peat bogs or lake sediments which are good sedimentary traps, but only the thickest ones are recorded

in other environment (*i.e.* “dry” outcrops).

Both lake catchments are covered by 4 - 6 m of Holocene sandy to silty deposits in which two major pumice layers are interbedded (Pl. 1A). These two pumice layers have been previously described. Naranjo & Moreno (1991) demonstrated that the lower pumice layer (*Llaima* pumice) was generated by *Llaima* Volcano (38° 42'S – 71° 44'W) at about 8830 ± 80 a BP (9900 ± 300 a cal BP) during a plinian eruption of its second eruptive phase called phase *Llaima* II. During this eruptive phase, a column of 4 km<sup>3</sup> of pumice reached around 40 km high and was dispersed by wind towards the south-east. This explosive stage ended with the collapse of the plinian column generating a 6-8 m thick base surge deposit. The Upper pumice layer (*Alpehué* pumice) took place during the last vigorous plinian eruption (column of 44 km high, 7.5 km<sup>3</sup> of airfall pumice and 0.4 km<sup>3</sup> of pyroclastic flow) of *Caldeira de Sollipulli* (38° 58'S – 71° 31'W) causing the formation of crater *Alpehué* at 2900 a BP (3020 ± 60 a cal BP) (Gilbert *et al.*, 1996; Naranjo *et al.*, 1993). In this short contribution, we document mineralogical data for



**Figure 1.** Schematic map of studied area. Black dots: sampled sites, OC1, OC2, OC3, OC4, CO1, CO2: outcrops; PB1 and PB2: peat bogs. The *Llaima* volcano is 40 km westwards.

the *Llaima* pumice. This provides some new insights into the mechanism of the eruption.

## 2. Material and method

### 2.1. Sample collection

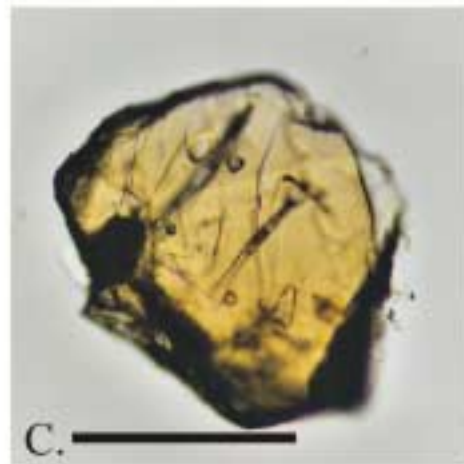
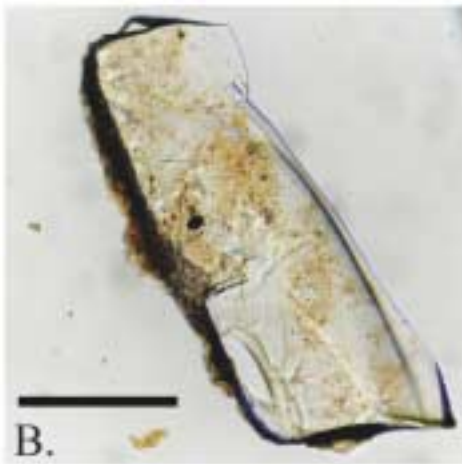
During the austral summer of 2002, a field campaign was conducted in the Icalma and Galletué watersheds. Five outcrops (e.g. Pl. 1.A) around the two lakes were sampled from the different watersheds (Fig. 1C). An additional outcrop was sampled westwards of the Icalma catchment (Fig. 1C, location CO2). Samples (around 1 dm<sup>3</sup> of bulk material) in each tephra bed were taken in respect of significant macroscopical changes of texture, grain size or colour occurs. Thus, the number and position of samples vary in the different studied outcrops.

Because of the dry summer climate, land reclamation (particularly for rearing), and high drainage (enhanced by the permeable nature of the volcanic layers), only the two lacustrine infilling peat bogs that subsist downstream

of Lake Galletué (Fig. 1) were cored. Coring was carried out with a Russian type steel corer (Belokopytkov & Beresnevich, 1955; Jowsey, 1965). These peat bogs are relatively heterogeneous and display alternating humid and dry zones. Thus, each peat bog was studied and pre-cored in order to determine the deepest site that could record the longest peat deposit.

### 2.2. Sample preparation and optical analyses

Outcrop samples containing particles coarser than 420 µm were roughly crushed and separated in four equal parts. An aliquot, made up of a small part of each quarter, was then re-crushed and sieved to isolate the grain size fraction between 105 and 420 µm. Samples containing only particles smaller than 420 µm were sieved without preliminary crushing to isolate the grain size fraction between 105 and 420 µm. In the cores from peat-bogs, the *Alpehué* pumice was visible to the naked eye (Pl. 1B). The *Llaima* pumice was not present, because these bogs are located outside the dispersion lobe. Each tephra bed was washed and treated in the same way as outcrop samples.



### Plate 1.

- A. View of outcrop OC3 (height: 6 m), the Llaima pumice is in the middle of the outcrop and the Alpehué pumice is at the top.
- B. Colourless olivine (Fo76-82).
- C. Honey yellow olivine (Fo32-35) from the Llaima pumice. Scale bar is 100  $\mu\text{m}$  long.

In all cases, smear slides were mounted as follows:

- (1) A small part of each 105–420  $\mu\text{m}$  fraction was mounted on a slide in Canada balsam and all the grains were determined in terms of their mineralogical nature.
- (2) Heavy minerals were separated by centrifuging 0.5  $\text{cm}^3$  of the 105–420  $\mu\text{m}$  fraction in test tubes containing 10 ml bromoform (density =  $2800 \text{ kg}\cdot\text{m}^{-3}$ ). Each tube's bottom was frozen in liquid nitrogen, allowing top the liquid containing the light fraction to be entirely discarded. The bromoform ice, containing the dense fraction was thawed, filtered and dried. Dense minerals were mounted on a slide. About 300 grains on each slides were identified and counted under polarising microscope using the 'Van Harten rubber technique' (Van Harten, 1965).

### 2.3. Chemical analyses

Monomineral spots from both pumice layers were isolated by hand-picking mineral grains in the 105–420  $\mu\text{m}$  fraction. All samples were mounted in PETREPOXY<sup>®</sup> resin. Major elements analyses were performed with a Cameca Camebax SX 50 electron microprobe from the C.I.A. of Louvain-la-Neuve University (Belgium). Analytical conditions are 15 kV, 10 nA current (analytical time: 10 seconds; bulk standard analyses used for calibration available from the author).

### 2.4. <sup>14</sup>C age dating

One sample of a charcoal-rich humus layer outcropping 15 cm above the *Llaima* pumice at location CO2 was collected for <sup>14</sup>C-dating. Dating was performed by A.M.S. (1.5 SDH-Pelletron Model "Compact Carbon AMS") at the Poznań Radiocarbon Laboratory (Poland). The sample was acid-base digested to remove any humic acid and fulvic acid, or any secondary carbonate deposited

in the sediment's organic fraction. It was then burned to obtain purified CO<sub>2</sub> for introduction into the spectrometer's carbon cathode. Calibrations were made according to Stuiver *et al.* (1998).

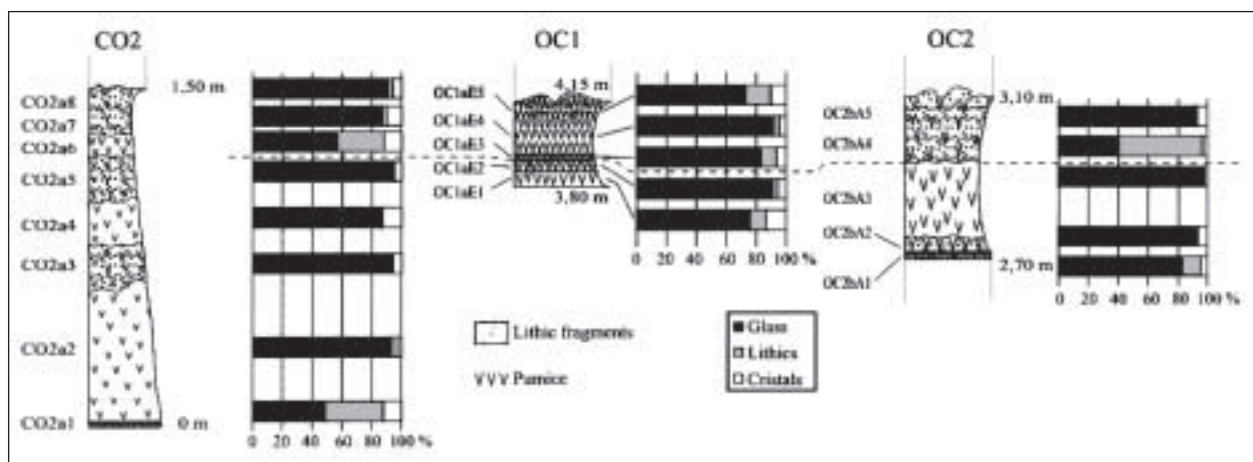
## 3. Results

### 3.1. <sup>14</sup>C age dating

The charcoal-rich humus was dated at  $9,030 \pm 45$  <sup>14</sup>C yr BP (Poz-252), corresponding to bracketed calibrated ages of  $10,180 \pm 70$  yr cal BP. Since our sample was taken just above the tephra, that result should be considered as a minimal age.

### 3.2. *Llaima pumice*

In the Icalma and Galletué catchments, the *Llaima* pumice is up to 100 cm thick. It is a yellowish to brownish coarse grained pumice layer with variable amounts of lithic fragments (1–39% in CO2, 3–17% in OC1, 0–57% in OC2; De Vleeschouwer, 2002). The pumice mineralogy is characterised by colourless glass shards, xenomorphic plagioclase, automorphic orthopyroxene and clinopyroxene, apatite, opaque minerals and in addition, a high amount of hypautomorphic to automorphic olivine (Pl. 1B–C). All these crystals are phenocrysts and their concentrations vary from 2 to 13% in respect to the different outcrops and samples. The olivine population can be divided into two groups based on colour: olivine phenocrysts are either colourless or honey-yellow. Glass shards are optically yellowish to brown, reflecting a relatively advanced degradation stage. These colours are darker at the deposit's base and top. This could be explained by a stagnation of percolating water at the deposit base, degrading the pumice more intensively.



**Figure 2.** Detailed logs and components for the *Llaima Pumice* sections CO2, OC1 and OC2. A fresh lithics outbreak is observed above the middle of the pumice deposit. Lithics are mainly composed of fragments of poorly differentiated magma (*i.e.* basaltic scoria *s.l.*) along with a small fraction of schists.

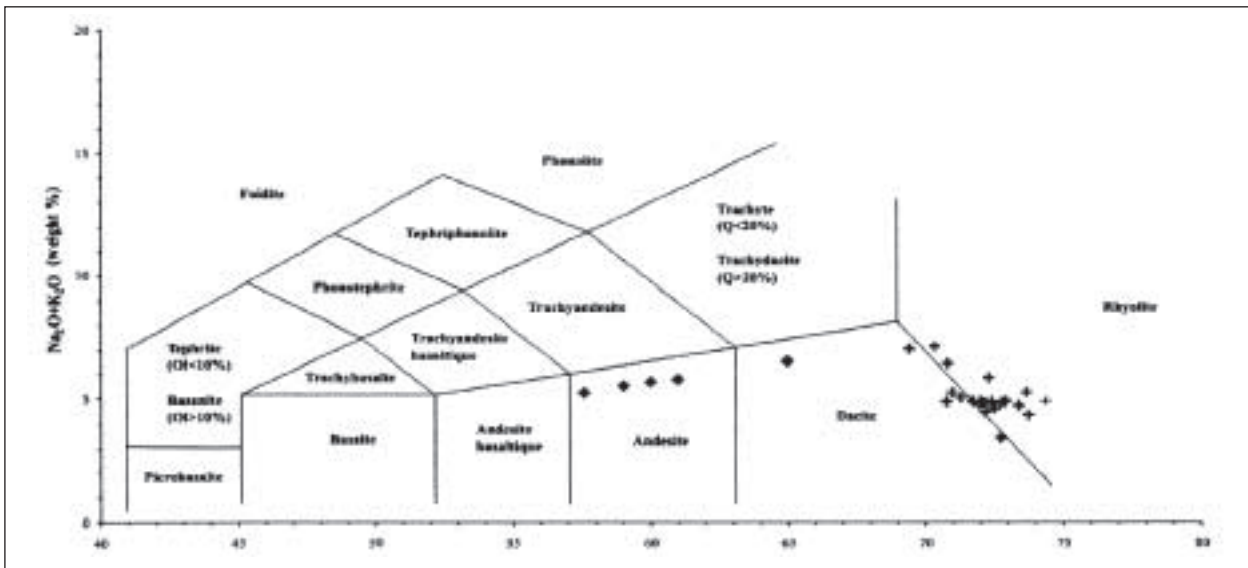


Figure 3. *Llama* Pumice glass shards: chemical analyses reported in the TAS vs.  $\text{SiO}_2$  diagram (Le Bas *et al.*, 1986); Black diamonds: whole rock analyses from the *Llama* pumice (Naranjo & Moreno, 1991)

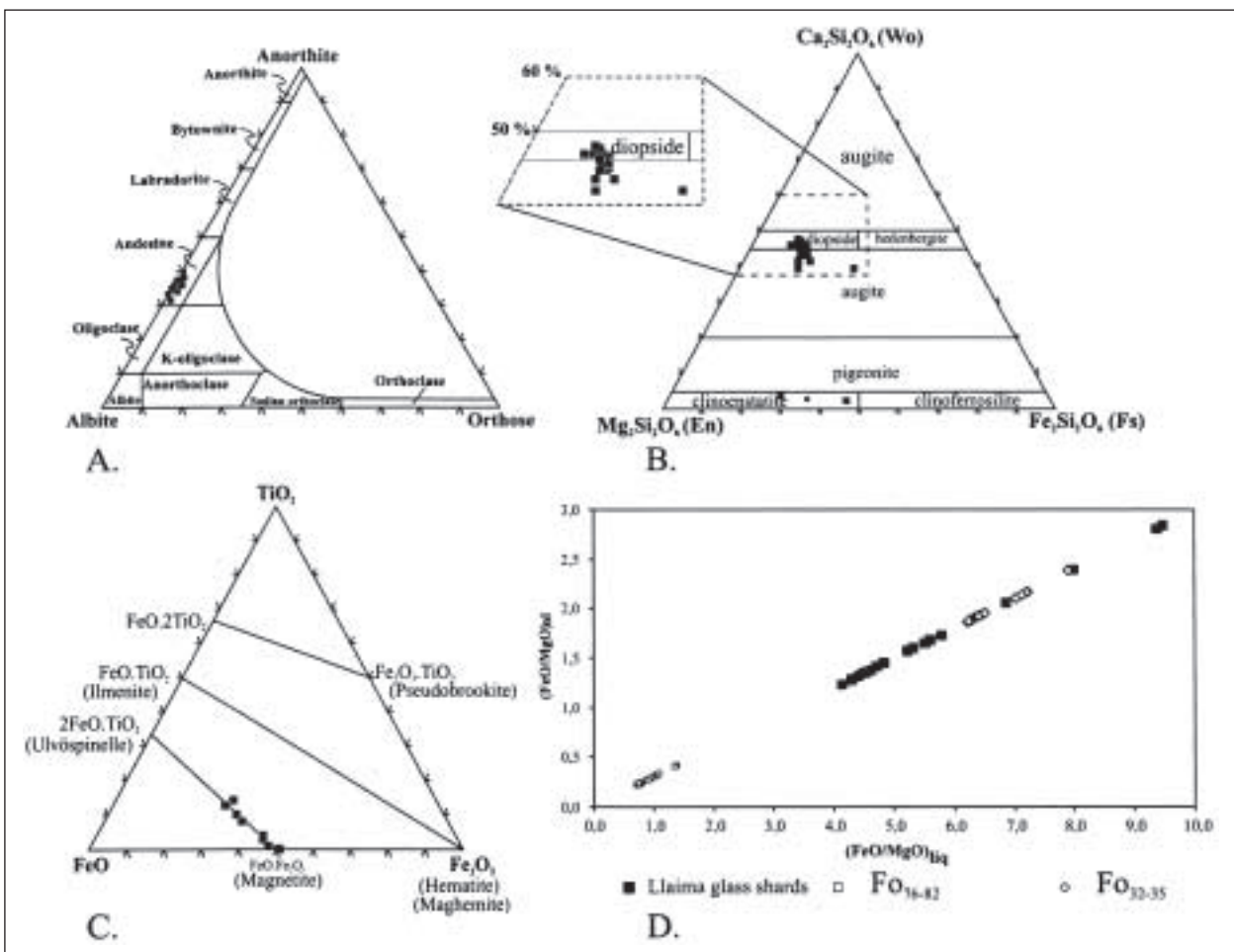


Figure 4. Chemical analyses of minerals found in the *Llama Pumice* reported in corresponding ternary diagrams. A. feldspars diagram. B. pyroxenes diagram according to Morimoto (1988). C. oxides diagram. D. Fe/Mg ratios diagram according to Roeder & Emslie (1970). Note that the  $\text{Fo}_{76-82}$  field is strongly isolated from the *Llama Pumice* glass shards' field.

Llaïma glass shards Mean (n=20)		Alpehué glass shards		Std dev
		Std dev	Mean (n=18)	
SiO <sub>2</sub>	72,14	1,24	74,88	0,27
TiO <sub>2</sub>	0,41	0,13	0,21	0,08
Al <sub>2</sub> O <sub>3</sub>	15,49	0,83	14,12	0,09
FeOt	3,42	0,56	1,50	0,12
Fe <sub>2</sub> O <sub>3</sub>	0,60	0,10	0,26	0,02
MgO	0,45	0,16	0,24	0,02
CaO	2,31	0,34	1,10	0,05
Na <sub>2</sub> O	3,28	1,01	3,26	0,77
K <sub>2</sub> O	1,84	0,21	4,24	0,10
<b>Total</b>	<b>99,94</b>		<b>99,81</b>	

	Plagioclase					Olivine (Fo <sub>32-35</sub> )	Olivine (Fo <sub>76-82</sub> )	
	Llaïma		Alpehué			Llaïma	Llaïma	
	Mean (n=12)	Std dev	Mean (n=10)	Std dev		Mean (n=19)	Mean (n=24)	Std dev
SiO <sub>2</sub>	58,11	4,14	58,44	1,77	SiO <sub>2</sub>	32,22	38,80	0,79
TiO <sub>2</sub>	0,00	0,00	0,00	0,00	TiO <sub>2</sub>	0,00	0,01	0,03
Al <sub>2</sub> O <sub>3</sub>	25,32	2,50	25,07	1,05	Al <sub>2</sub> O <sub>3</sub>	0,00	0,01	0,02
FeO	0,32	0,13	0,29	0,07	FeO	50,61	18,99	2,93
MnO	-	-	-	-	MnO	1,00	0,10	0,17
MgO	0,02	0,02	0,01	0,01	MgO	14,41	41,09	2,22
CaO	8,19	3,07	7,79	1,20	CaO	0,20	0,21	0,03
Na <sub>2</sub> O	6,74	1,75	6,84	0,64	Na <sub>2</sub> O	0,00	0,00	0,01
K <sub>2</sub> O	0,17	0,06	0,42	0,12	K <sub>2</sub> O	0,00	0,00	0,01
Cr <sub>2</sub> O <sub>3</sub>	-	-	-	-	Cr <sub>2</sub> O <sub>3</sub>	0,00	0,00	0,00
<b>Total</b>	<b>98,86</b>	<b>0,45</b>	<b>98,95</b>	<b>0,30</b>	<b>Total</b>	<b>98,45</b>	<b>99,21</b>	<b>0,56</b>
<b>Cationic proportions calculated on a five cations basis</b>					<b>Cationic proportions calculated on a three cations basis</b>			
Si <sup>4+</sup>	2,63	0,16	2,65	0,07	Si	1,00	1,00	0,01
Ti <sup>4+</sup>	0,00	0,00	0,00	0,00	Ti	0,00	0,00	0,00
Al <sup>3+</sup>	1,37	0,16	1,35	0,07	Al <sup>3+</sup>	0,00	0,00	0,00
Fe <sup>3+</sup>	0,01	0,01	0,01	0,00	Fe <sup>3+</sup>	0,00	0,00	0,00
Mn <sup>2+</sup>	-	-	-	-	Fe <sup>2+</sup>	1,31	0,41	0,07
Mg <sup>2+</sup>	0,00	0,00	0,00	0,00	Mn <sup>2+</sup>	0,03	0,00	0,00
Ca <sup>2+</sup>	0,40	0,16	0,38	0,06	Mg <sup>2+</sup>	0,66	1,58	0,07
Na <sup>+</sup>	0,59	0,15	0,60	0,05	Ca <sup>2+</sup>	0,01	0,01	0,00
K <sup>+</sup>	0,00	0,00	0,02	0,01	Na <sup>+</sup>	0,00	0,00	0,00
Cr <sup>3+</sup>	-	-	-	-	K <sup>+</sup>	0,00	0,00	0,00
					Cr <sup>3+</sup>	0,00	0,00	0,00
Ab	59,16	15,19	59,86	5,47	Fo	32,42	79,29	3,52
An	39,88	15,51	37,72	5,88				
Or	0,96	0,37	2,42	0,72				

	Orthopyroxene				Clinopyroxene			
	Llaima		Alpehué		Llaima		Alpehué	
	Mean (n=2)	Std dev	Mean (n=5)	Std dev	Mean (n=26)	Std dev	Mean (n=5)	Std dev
SiO <sub>2</sub>	51,81	1,53	52,28	0,47	50,26	0,80	52,06	0,22
TiO <sub>2</sub>	0,35	0,02	0,18	0,14	0,64	0,13	0,26	0,06
Al <sub>2</sub> O <sub>3</sub>	0,92	0,49	0,54	0,23	2,76	0,80	0,85	0,16
FeO	22,19	6,00	24,74	1,44	8,62	2,03	11,86	0,23
MnO	0,29	0,40			0,28	0,17		
MgO	21,32	4,45	20,04	1,18	14,68	0,91	13,28	0,23
CaO	1,50	0,40	1,34	0,06	21,05	1,04	20,58	0,24
Na <sub>2</sub> O	0,00	0,00	0,00	0,00	0,29	0,04	0,33	0,03
K <sub>2</sub> O	0,00	0,00	0,00	0,00	0,00	0,01	0,00	0,00
Cr <sub>2</sub> O <sub>3</sub>	0,00	0,00			0,08	0,15		
<b>Total</b>	<b>98,37</b>	<b>1,27</b>	<b>99,11</b>	<b>0,66</b>	<b>98,67</b>	<b>0,51</b>	<b>99,21</b>	<b>0,21</b>
<b>Cationic proportions calculated on a four cations basis</b>								
Si <sup>4+</sup>	1,97	0,03	1,99	0,02	1,89	0,03	1,97	0,01
Ti <sup>4+</sup>	0,01	0,00	0,01	0,00	0,02	0,00	0,01	0,00
Al <sup>3+</sup> (IV)	0,03	0,03	0,01	0,02	0,12	0,04	0,03	0,01
Al <sup>3+</sup> (VI)	0,01	0,01	0,01	0,01	0,00	0,00	0,01	0,01
Fe <sup>3+</sup>	0,01	0,02	0,01	0,01	0,09	0,03	0,03	0,02
Fe <sup>2+</sup>	0,70	0,24	0,78	0,05	0,19	0,08	0,35	0,01
Mn <sup>2+</sup>	0,01	0,01			0,01	0,01		
Mg <sup>2+</sup>	1,20	0,20	1,14	0,05	0,82	0,05	0,75	0,01
Ca <sup>2+</sup>	0,06	0,01	0,05	0,00	0,85	0,04	0,83	0,01
Na <sup>+</sup>	0,00	0,00	0,00	0,00	0,02	0,00	0,02	0,00
K <sup>+</sup>	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00
Cr <sup>3+</sup>	0,00	0,00			0,00	0,00		

**Table 1.** Mean values and cationic proportions of chemical analyses for glass shards and minerals from the *Llaima* pumice (the *Alpehué* pumice is given for comparison).

The degradation of the deposit top can be accounted for meteoritic weathering of the tephra. A fresh outbreak of lithics unaccompanied by any erosion structure is observed in the pumice at three locations. In fact, lithics concentration increases from 1 to 32%, 115 cm above the base of the pumice at CO2; from 3 to 11%, 10 cm above the base of the pumice at OC1, and from 0 to 57%, 25 cm above the base of the pumice at OC2 (Fig. 2). Since the increase in lithic content is abrupt, it may reflect a structural change in the source vent as the explosions blew off older rock fragments from the partially collapsed crater and upper conduit. In such context, Llaima pumice can be considered as deposited in two pulses during a single plinian eruption phase.

*Llaima* pumice has an andesitic to dacitic composition (60–65% SiO<sub>2</sub>), as already shown by Naranjo & Moreno (1991). Glass shards are rhyodacitic (70–74% SiO<sub>2</sub>; Fig. 3),

transparent minerals are made of andesine (Fig. 4A) and fluorapatite. Neither K-feldspar nor quartz was detected. Orthopyroxene is hypersthene and clinopyroxenes are made of Ca-augite and diopside (Fig. 4B). Opaque minerals are titanomagnetites (Fig. 4C). The transparent olivine is made of Fo<sub>76</sub> to Fo<sub>82</sub> whereas the honey yellow one is made of Fo<sub>32</sub> to Fo<sub>35</sub>. Mean chemical analyses and cationic proportions are reported in Table 1.

## 4. Discussion

### 4.1. Age of the eruption

In spite of the fact that the age obtained in this work (10,180 ± 70 yr cal BP) is a minimal age, it is consistent with the age obtained by Naranjo & Moreno (1991): 9900

$\pm 300$  yr cal BP for a charcoal taken above the pumice layer. Both results place the tephra-fall around 10,000 yr cal BP. However, Mardones *et al.* (1993) have dated a silty to clayey sample that was taken just below a pumice tephra layer interpreted as the equivalent of *Llaima* pumice in a core of a nearby peat bog (Guallemapu, S 38°72'–W 71°13', pumice depth between 95 and 105 cm, sample depth: 125 cm). It has given an age of  $4580 \pm 70$  yr BP ( $5225 \pm 275$  yr cal BP). Provided the tephra correlation is accepted, such a difference between Mardones *et al.* (1993) and our dating cannot be reconciled. Mineralogical, chemical and age consistencies between Naranjo & Moreno work (1991) and ours implies that the Mardones *et al.* (*op. cit.*) date is not useable, probably because of a young carbon contamination by water percolation or presence of rootlets through the pumice deposit.

#### 4.2. Eruption process

The key founding of the present study is the occurrence of two different olivine populations in the same tephra as well as olivine in rhyodacitic liquids. Olivine had already been optically identified in the *Llaima* pumice by Naranjo & Moreno (1991). Our investigation shows that these olivine populations are optically but also chemically different. The Fe/Mg ratio for both olivine populations and for the *Llaima* pumice's glass shards is reported on Fig. 4D using the terminology Roeder & Emslie (1970). The  $Fo_{32-35}$  field is overlapped by the glass shard's one. The  $Fo_{76-82}$  is isolated. Both observations demonstrate that  $Fo_{32-35}$  crystallised in equilibrium with the magma and that  $Fo_{76-82}$  is in disequilibrium. This is consistent with the fact that even if olivine is more common in basic magmas, Fe-rich olivine is known to be a common mineral phase in the dacitic liquid from this region (Futa & Stern, 1988; López-Escobar, 1984). Moreover, olivine with bimodal distributions ( $Fo_{24-26}$  and  $Fo_{74-87}$ ) have already been observed in andesites from Puyehue volcano (Gerlach *et al.*, 1988). These andesites from Puyehue volcano are interpreted to be disequilibrium phenocrysts resulted from a mixing between a basaltic magma and an evolved magma to produce supercooled mixtures. The structural study of the *Llaima* pumice had already suggested a mixing with a more basic (*i.e.* hotter) magma at the bottom of the *Llaima* magmatic chamber (Naranjo & Moreno, 1991). The presence of  $Fo_{76-82}$  is consistent with this hypothesis. The eruption of the *Llaima* pumice was caused by mixing between a resting acid magma with a replenishment batch of more basic magma containing  $Fo_{76-82}$ .

#### 5. Summary and conclusion

Two major tephra blanketed the catchments of lakes Icalma and Galletué (southern Chile) during the Holocene. The lower one, the *Llaima pumice* (Naranjo & Moreno, 1991), is an olivine-bearing andesite to dacite

with pyroxene and titanomagnetite. It erupted in two pulses of a single eruption phase, from Volcano *Llaima* during the early Holocene ( $10,180 \pm 70$  yr cal BP – this work,  $9900 \pm 300$  yr cal BP – Naranjo & Moreno, 1991). The eruption was triggered by magma mixing as suggested in the previous study of Naranjo & Moreno (*op. cit.*) and by the two populations of completely different compositions of olivine, one of which did not crystallise in equilibrium with the magmatic liquid.

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