Aerosol trace element solubility determined using ultrapure water batch leaching: an intercomparison study of four different leaching protocols

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Abstract

Solubility of aerosol trace elements, which determines their bioavailability and reactivity, is operationally defined and strongly depends on the leaching protocol used. Ultrapure water batch leaching is one of the most widely used leaching protocols, while the specific leaching protocols used in different labs can still differ in agitation methods, contact time, and filter pore size. It is yet unclear to which extent the difference in these experimental parameters would affect the aerosol trace element solubility reported. This work examined the effects of agitation methods, filter pore size and contact time on the solubility of nine aerosol trace elements, and found that the difference in agitation methods (shaking vs. sonication), filter pore size (0.22 vs. 0.45 μm), and contact time (1 vs. 2 h) only led to small and sometimes insignificant difference in the reported solubility. We further compared aerosol trace element solubility determined using four ultrapure water leaching protocols which are adopted by four different labs and vary in agitation methods, filter pore size and/or contact time, and observed good agreement in the reported solubility. Therefore, our work suggests that although ultrapure water batch leaching protocols used by different labs vary in specific experimental parameters, the determined aerosol trace element solubility is comparable. We recommend ultrapure water batch leaching to be one of the reference leaching schemes, and emphasize that additional consensus in the community on agitation methods, contact time and filter pore size is needed to formulate a standard operating procedure for ultrapure water batch leaching.

1 Introduction

Aerosol trace elements, originating from natural and anthropogenic sources, are of great concerns, as they significantly impact marine and terrestrial ecosystems (Boyd and Ellwood, 2010; Dong et al., 2023; Mahowald et al., 2018), have adverse effects on human health (Dahmardeh Behrooz et al., 2021; Fang et al., 2017; Gao et al., 2022; Wei et al., 2019), and play important roles in atmospheric chemistry (Al-Abadleh, 2024; Alexander et al., 2009; Mao et al., 2013; Martin and Hill, 1987; Wang et al., 2021). The dissolved fraction of aerosol trace elements, instead of their total abundance, is considered to be bioavailable (Baker and Croot, 2010; Ito et al., 2012; Mukhtar and Limbeck, 2013) and more chemically reactive in the atmosphere (Kebede et al., 2016; Mao et al., 2017). Dissolved trace elements are typically referred to as the fraction of elements which can pass through a filter with certain pore size (usually 0.2-0.22 or 0.45 μm) after aerosol particles are dissolved in certain aqueous solutions (Boyd and Ellwood, 2010; Ito and Xu, 2014; Meskhidze et al., 2016; Myriokefalitakis et al., 2018). Solubility (or fractional solubility, to be more precise), which is defined as the ratio (in %) of the dissolved element to the total element (Baker et al., 2006; Sholkovitz et al., 2012), largely determines the bioavailability and reactivity of aerosol trace elements.

A wide range in the solubility has been reported in the literature for a given trace element in atmospheric aerosols, and for example, the reported solubility of aerosol Fe ranges from <1% to >90% (Baker and Jickells, 2006; Sholkovitz et al., 2012). Such wide variabilities in aerosol trace element solubility, on one hand, can be caused by difference in sources and aging processes of aerosol particles examined (Ito et al., 2021; Meskhidze et al., 2019); on the other hand, they also stem from various leaching protocols which were used by different studies

66   (Chen et al., 2006; Li et al., 2023; Upadhyay et al., 2011).

67   Various leaching protocols have been used in previous studies to extract dissolved aerosol

68   trace elements, as summarized in a recent paper (Li et al., 2023). In brief, available leaching

69   protocols broadly consist of two catalogues, including flow-through leaching and batch

70   leaching. Flow-through leaching is instantaneous and typically has a contact time (between

71   aerosol particles and the leaching solution used) of tens of seconds, and batch leaching usually

72   has a much longer contact time (tens of minutes or longer). Compared to flow-through leaching,

73   batch leaching is more widely used in atmospheric research. Furthermore, for batch leaching,

74   various leaching solutions were used in previous studies, such as ultrapure water, filtered

75   seawater, and formate/acetate buffers. Compared to filtered seawater and formate/acetate

76   buffers, ultrapure water is more widely used in atmospheric research due to its simplicity and

77   reproducibility (Li et al., 2023; Meskhidze et al., 2019); another important reason is that

78   ultrapure water leaching does not introduce any other chemical species (except water) and thus

79   can simultaneously extract water-soluble ions and organics for additional analysis.

80   Even for ultrapure water batch leaching, protocols used by different studies may still vary

81   in agitation methods, contact time, and filter pore size; nevertheless, the effects of these factors

82   on the reported solubility are not well understood. First, some labs use sonication to agitate the

83   leaching solutions (Chen et al., 2006; Kumar and Sarin, 2010; Liu et al., 2021; Longo et al.,

84   2016; Shi et al., 2020), and other labs use shaking (Baker et al., 2003; Gao et al., 2020; Hsu et

85   al., 2010; Li et al., 2022; Salazar et al., 2020). Sonication may cause changes in chemical

86   composition and formation of reactive oxygen species in the solution (Juretic et al., 2015;

87   Miljevic et al., 2014); however, it remains to be examined whether sonication will change the

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88   solubility of aerosol trace elements. Second, filters with different pore sizes, including 0.2-0.22

89   and 0.45 μm (and 0.02 μm to a less extent), are employed to filter the leaching solutions,

90   contributing to the uncertainties in the reported solubility; however, the effects of filter pore

91   size have seldom been experimentally examined. Third, some studies (Li et al., 2023; Mackey

92   et al., 2015) suggested that contact time (2-8 h) could also influence the reported solubility.

93   In the present work, using aerosol particles collected at a suburban site close to the

94   coastline of the Northwest Pacific, we investigated to which extent different ultrapure water

95   batch leaching protocols would affect reported aerosol trace element solubility. In the first part

96   of this work, we examined the effects of agitation (shaking vs. sonication), filter pore size (0.22

97   vs. 0.45 μm) and contact time (1 vs. 2 h) on the reported solubility of nine aerosol trace elements.

98   In the second part, we compared solubility determined using protocols commonly adopted by

99   four labs. The four labs all use ultrapure water batch leaching, but the leaching protocols they

100   use differ in agitation method, contact time, and/or filter pore size.

101   **2 Experimental section**

102   **2.1 Sample collection and distribution**

103   We collected aerosol samples between 18 March and 22 April 2023 in Qingdao, a coastal

104   city in northern China, typically impacted by desert dust and anthropogenic aerosols in spring.

105   As described elsewhere (Zhang et al., 2022), aerosol sampling took place at a suburban site

106   which was about 1.3 km from the coast. A cumstom-made high-volume aerosol sampler (ASM-

107   1; flow rate: 1 m^3/min) was deployed on a building roof (about 20 m above the ground) to

108   collect PM_{10} samples. Aerosol sampling started at 08:00 am each day and stopped at 07:30 am

109   on the next day, resulting in a sampling volume of 1410 m^3. PM_{10} samples were collected onto

110   pre-cleaned Whatman 41 cellulose fiber filters (25 cm × 20 cm) which had very low

111   backgrounds for trace elements (Morton et al., 2013; Zhang et al., 2022). In total we collected

112   26 filter samples, 4 sampling blanks, and 3 lab blanks: lab blanks were defined as pre-cleaned

113   filters, and sampling blanks were defined as pre-cleaned filters which were placed in the aerosol

114   sampler for 2 h when the sampling flow was off. The amounts of dissolved trace elements on

115   blank filters were mostly below detection limits; in a few cases the blank levels exceeded

116   detection limits, but were negligible when compared to these on filter samples.

117   After aerosol collection, each filter was folded inward and placed into a clean

118   polyethylene bag (12 inch × 9 inch, supplied by Sigma-Aldrich) which was used due to its low

119   background (Morton et al., 2013), and then stored at -20 °C. A titanium punch was used to

120   obtain 10 subsamples (47 mm in diameter) from each filter sample, and these subsamples were

121   stored at -20 °C.

2.2 Measurement of total and dissolved trace elements

2.2.1 Total elements

124   As shown in Table 1 and described below, for the 10 subsamples obtained from each

125   original filter sample, the first subsample was digested to determine total elements, another

126   eight subsamples were leached using different protocols to determine dissolved elements, and

127   the last subsample was reserved for any unforeseen purpose (but was not used at the end).

128   Subsample 1 was digested in a Teflon jar which contained a mixture of HNO_3-HF-H_2O_2,

129   using a microwave digestion system (Zhang et al., 2022). After digestion, we evaporated

130   residual acids in the Teflon jar, and filled it with 20 mL HNO_3 (1%). Subsequently, we filtered

131   the solution through a polyethersulfone membrane syringe filter (with a pore size of 0.22 μm),

and then used ICP-MS (inductively coupled plasma mass spectrometry, iCAP Q, Thermo

Fisher) to measure nine trace elements, including Fe, Al, As, Cr, Cu, Mn, Pb, V and Zn. These

elements were chosen because they are important nutrients, toxic elements, or source tracers.

Table 1. Overview of protocols used to digest and leach subsamples examined in this work.
For each protocol, 26 subsamples were examined. In this table, "Lab" represents the lab whose
protocol was adopted in this work to digest or leach subsamples. Experimental parameters for
subsamples 2c-2e, when different from those for subsamples 2a, are highlighted in bold and
underline.

subsample	agitation	contact time (h)	filter pore size (μm)	Lab	References
1	digestion	--	--	GIG	Zhang et al. (2022)
2a	shaking	2	0.22	GIG	Zhang et al. (2022)
2b	shaking	2	0.22	GIG	--
2c	shaking	2	**0.45**	--	--
2d	**sonication**	2	0.22	--	--
2e	shaking	**1**	0.22	--	--
3a	sonication	1	0.22	ZJU	Liu et al. (2021)
3b	sonication	1	0.45	OUC	Shi et al. (2020)
3c	sonication	0.5	0.20	NIO	Panda et al. (2022)

2.2.2 Dissolved elements

Subsample 2a was leached using the protocol adopted by the lab at Guangzhou Institute

of Geochemistry (GIG) (Li et al., 2023; Zhang et al., 2022; Zhang et al., 2023). In brief, the

subsample was shredded and then immersed in 20 mL ultrapure water for 2 h, stirred using an

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146 orbital shaker; subsequently, the solution was filtered through a polyethersulfone membrane

147 syringe filter (with a pore size of 0.22 μm). After that, the solution was immediately acidified

148 with a small volume of high-purity HNO_3 to contain 1% HNO_3, and then analyzed using ICP-

149 MS to determine dissolved trace elements. Subsample 2b was leached using the same protocol

150 as subsample 2a, and the purpose was to examine whether aerosol particles were

151 homogeneously distributed on different subsamples, and to assess the repeatability of the GIG

152 leaching protocol.

153 Subsamples 2c-2e were leached using protocols similar to that used to leach subsample

154 2a. As summarized in Table 1, the only difference to the protocol used to leach subsample 2a

155 was the filter pore for 2c (0.45 μm, vs. 0.22 μm for 2a), agitation method for 2d (sonication, vs.

156 shaking for 2a), and contact time for 2e (1 h, vs. 2 h for 2a). The purpose of using subsamples

157 2c-2e is to examine the effects of filter pore size (0.22 vs. 0.45 μm), agitation method (shaking

158 vs. agitation), and contact time (1 vs. 2 h) on the reported solubility.

159 Subsamples 3a, 3b and 3c were leached using the protocols typically used by ZJU

160 (Zhejiang University, China), OUC (Ocean University of China, China) and NIO (National

161 Institute of Oceanography, India), respectively, in order to compare solubility determined by

162 the GIG lab with those reported by the other three labs. Please note that subsamples 3c were

163 leached and analyzed by NIO, while subsamples 3b and 3c were leached and analyzed at GIG

164 (using the ZJU and OUC protocols, respectively).

165 Subsample 3a was leached at GIG using the ZJU protocol (Liu et al., 2021; Zhu et al.,

166 2020). In brief, each subsample was shredded and immersed in 20 mL ultrapure water, and the

167 aqueous mixture was sonicated for 1 h during which the water bath temperature was kept below

168   30 °C; after that, the aqueous mixture was filtered using a polyethersulfone membrane syringe

169   filter (pore size: 0.22 μm) and acidified for later ICP-MS analysis. Subsample 3b was leached

170   at GIG using the OUC protocol (Shi et al., 2020), which is very similar to the ZJU method: the

171   only difference is that the filter pore size was 0.45 μm for the OUC protocol and 0.22 μm for

172   the ZJU protocol.

173   Subsample 3c was leached and analyzed at NIO using the NIO protocol (Panda et al.,

174   2022). In brief, each subsample was shredded and placed into a pre-cleaned Savillex vial (50

175   mL); after that, the vial was filled with 20 mL ultrapure water, caped, and then sonicated for

176   30 minutes to agitate the aqueous mixture (but in 2 cycles with 15 min for each cycle, in order

177   to maintain the water bath at room temperature). The aqueous mixture was then filtered through

178   a Whatman PVDF syringe filter (pore size: 0.2 μm), and then acidified with HNO_3 (2% v/v)

179   for later high-resolution ICP-MS analysis (Nu Instruments, Attom ES).

3 Results and discussion

181   Subsamples 2a and 2b were identically leached using the protocol GIG normally uses, and

182   the paired *t*-test (α = 0.05) was employed to examine whether the difference in obtained

183   solubility was significant. As summarized in Table 2, the difference in obtained solubility was

184   not statistically significant between 2a and 2b for Fe, Al, As, Mn, Pb, and V; furthermore,

185   Figure S1 suggests good linear correlations in solubility between 2a and 2b for the six elements

186   (*r* > 0.99), and the corresponding slopes (0.98 to 1.02) were very close to 1. For the other three

187   elements (Cr, Cu and Zn), although the difference in solubility was found to be statistically

188   significant between 2a and 2b (Table 1), good linear correlations between the solubility were

189   found (*r* > 0.97) and the slopes (1.00-1.06) were close to 1; therefore, the difference in solubility

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190 between 2a and 2b, if it existed, was small for Cr, Cu and Zn.

191 In summary, we conclude that the distribution of aerosol particles on a given original filter

192 was homogeneous and that the protocol GIG normally uses had very good repeatability.

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194 **Table 2.** Summary of statistical analysis (paired-t-test, $\alpha = 0.05$) which examined whether the

195 difference in solubility obtained for different groups of subsamples is statistically significant.

196 Solubility obtained for subsamples 2a is compared with those obtained for subsamples 2b, 2c,

197 2d and 2e, respectively. Y: the difference is statistically different; N: the difference is not

198 statistically different.

element	2a vs. 2b	2a vs. 2c	2a vs. 2d	2a vs. 2e
Fe	N	**Y**	**Y**	**Y**
Al	N	N	N	**Y**
As	N	N	**Y**	**Y**
Cr	**Y**	**Y**	**Y**	**Y**
Cu	**Y**	**Y**	**Y**	**Y**
Mn	N	N	**Y**	**Y**
Pb	N	**Y**	**Y**	N
V	N	N	N	N
Zn	**Y**	**Y**	**Y**	**Y**

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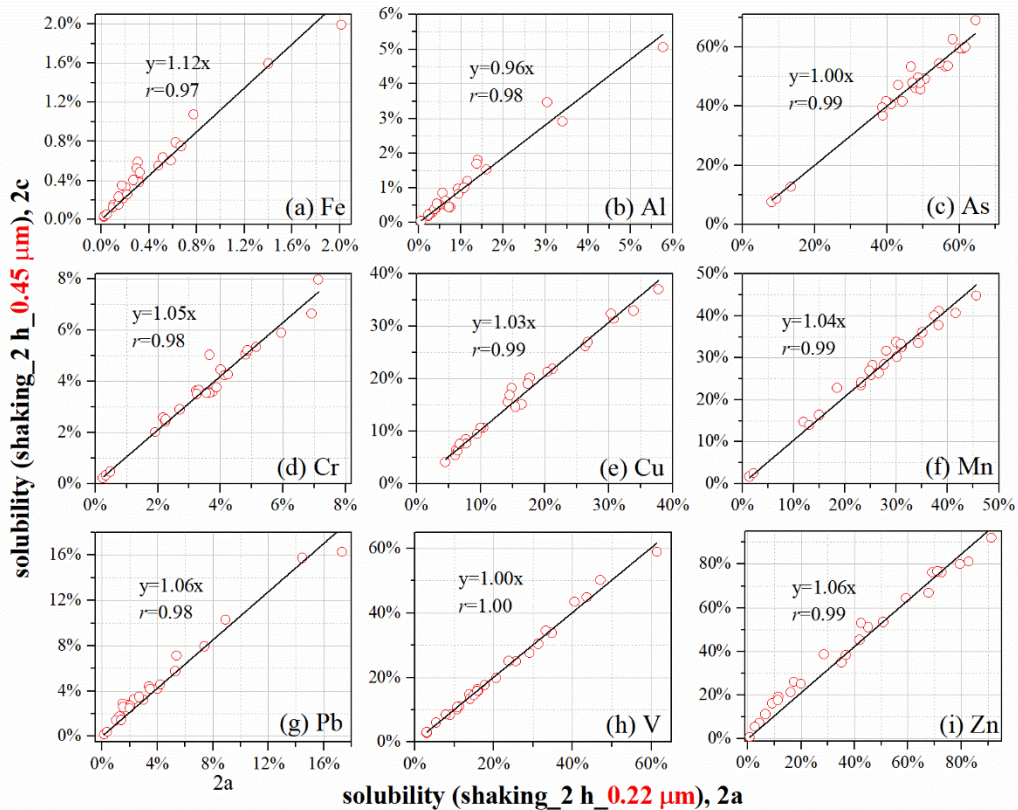## 3.1 The effects of filter pore size

201 To examine the effects of filter pore size on the reported solubility, subsamples 2a and 2c

202 were leached using very similar protocols, and the only difference is the pore size (2a: 0.22 μm;

203 2c: 0.45 μm) of filters used (Table 1).

204 The difference in obtained solubility was not statistically significant between 2a and 2c

205   for Al, As, Mn and V (Table 2); moreover, good linear correlations between 2a and 2c were

206   found for the four elements (Figure 1), with slopes (0.96-1.04) very close to 1. For the other

207   five elements (Fe, Cr, Cu Pb, and Zn), the difference in solubility between 2a and 2c was found

208   to be statistically significant; however, solubility between 2a and 2c was very well linearly

209   correlated (Figure 1), with slopes (1.03-1.12) close to or slightly larger than 1.

210   To conclude, among the nine elements we examined, the effects of filter pore size (0.22

211   vs. 0.45 μm) on reported solubility were found to be insignificant for four elements (Al, As,

212   Mn and V) and very smaller for the other five elements (Fe, Cr, Cu, Pb and Zn).

213

214   **Figure 1.** The effects of filter pore size (2a: 0.22 μm; 2c: 0.45 μm) on measured element

215   solubility. The only difference in protocols used to leach subsamples 2a and 2c is the filter pore

216   size (0.22 versus 0.45 μm).

217

3.2 The effects of agitation

As shown in Table 1, the only difference between the protocol used to leach subsamples 2a and that used to leach subsamples 2d is the agitation method used (2a: shaking; 2d: sonication), and solubility obtained for subsamples 2a and 2d was compared to assess the effects of agitation methods on reported solubility.

Table 2 shows that the reported solubility between 2a and 2d was not statistically different for two elements (Al and V); in addition, good linear correlations between 2a and 2d were found for the two elements (Figure 2), and these slopes (1.10 for Al and 1.05 for V) were quite close to 1. With respect to the other seven elements (Fe, As, Cr, Cu, Mn, Pb and Zn), on one hand, the difference in solubility between 2a and 2d was found to be statistically significant; on the other hand, good linear correlations in solubility existed between 2a and 2d (Figure 2), and these slopes were in the range of 0.94-1.21.
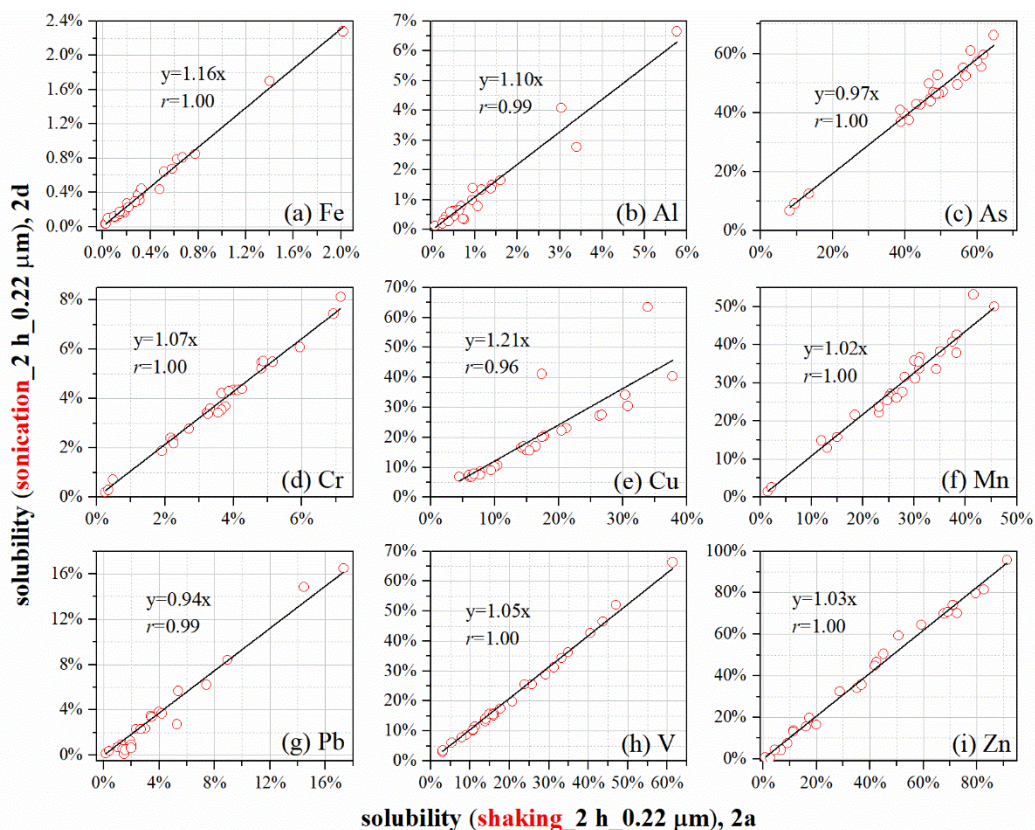
Figure 2. The effects of agitation (2a: shaking; 2d: sonication) on measured element solubility. The only difference in protocols used to leach subsamples 2a and 2d is the agitation method (shaking vs. sonication).

In summary, we found that the choice of agitation methods (shaking vs. sonication) had no measurable (for Al and V) or small effects (for Fe, As, Cr, Cu, Mn, Pb and Zn) on the reported solubility.

3.3 The effects of contact time

To assess the impacts of contact time on reported solubility, subsamples 2a and 2e were leached using very similar protocols, and the only difference was contact time (2a: 2 h; 2e: 1 h). We examined the effects of these two contact time, as the contact time was 2 h for the GIG protocol, and 0.5-1 h for ZJU, OUC and NIO protocols (Table 1).

As shown in Table 2, the reported solubility were not statistically different between 2a and 2e for Pb and V; moreover, good linear relationships between 2a and 2e were found for the two elements (Figure 3), with slopes close to 1 (1.24 for Pb and 1.02 for V). For the other seven elements, their solubility was found to be statistically significant between 2a and 2e (Table 2); nevertheless, for each of the seven elements, the solubility reported for 2a was very well linearly correlated with that reported for 2e (Figure 3), and the slopes were close to 1 (in the range of 0.95-1.28).
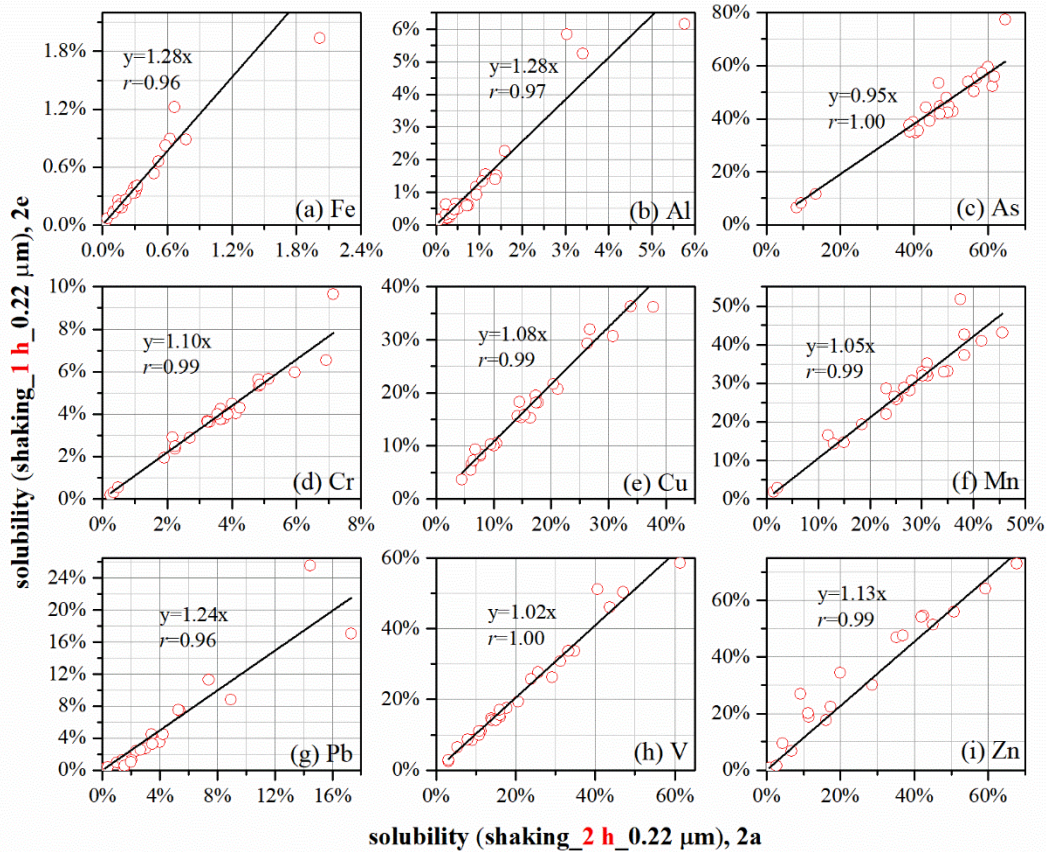
Figure 3. The effects of contact time (2a: 1 h; 2e: 2 h) on measured element solubility. The only difference in protocols used to leach subsamples 2a and 2e is the contact time (1 h versus 2 h).

To summarize, our present work suggested that the increase in contact time from 1 h to 2 h would cause insignificant or small effects on reported solubility. Using a different set of aerosol samples, our previous work (Li et al., 2023) compared the measured solubility obtained with longer contact time (4 and 8 h) to that obtained with a contact time of 2 h. As shown in Table S1, increase in contact time from 2 to 4 h would cause significant increase in solubility, on average by a factor of ~1.3 for Zn to ~3.1 for As (Li et al., 2023). It is still not clear why the increase in contact time from 1 to 2 h would not cause significant change in aerosol trace element solubility while the increase in contact time from 2 to 4 h would, probably because for

263   a given element, different speciation have different dissolution kinetics.

264   **3.4 Comparison of solubility obtained using protocols commonly used by four labs**

265   We further compared solubility determined using the GIG protocol with those determined

266   using ZJU, OUC and NIO protocols, respectively. Table 3 summarizes the slopes obtained from

267   correlation analysis (Figures 4 and S2-S8). The NIO lab measured eleven elements, among

268   which five elements (Fe, Al, Cu, Mn, and V) were measured using the other three protocols; as

269   a result, the solubility of these five elements determined using the NIO protocol was compared

270   with those determined using the GIG protocol.

271

272   **Table 3.** Correlations between solubility determined using the GIG protocol and that

273   determined using ZJU, OUC and NIO protocols. Here only the slopes (k) are provided.

|	ZJU	OUC	NIO
	k	k	k
Fe	1.03	1.39	1.82
Al	1.09	1.19	1.80
As	0.96	0.87	--
Cr	0.96	0.97	--
Cu	1.06	1.04	0.99
Mn	0.99	0.98	1.09
Pb	0.97	1.01	--
V	1.06	0.99	1.05
Zn	0.97	0.94	--

274

275   With respect to Fe solubility, GIG results were very well correlated with ZJU results ($r =$

276   0.99, Figure 4a) and the slope was found to be 1.03, suggesting good agreement between GIG

277   and ZJU; GIG results were also well correlated with while overall larger than OUC results (r

278   $= 0.98$, Figure 4b), and the slope was determined to be 1.39; good correlation was also found

279   between GIG and NIO results ($r = 0.91$, Figure 4c), and the slope was determined to be 1.82,

280   indicating that Fe solubility determined using the NIO protocol was larger than that determined

281   using the GIG protocol. Similarly, with respect to Al solubility (Figure S2 and Table 3), the

282   GIG results were well correlated with ZJU, OUC and NIO results, and correlations were best

283   for ZJU ($r = 0.99$) and moderate for NIO ($r = 0.93$); in addition, the slopes were determined to

284   be 1.09, 1.19 and 1.80 for ZJU, OUC and NIO results, respectively.

285   With respect to Cu (Figure S5), Mn (Figures 4d-4f) and V (Figure S7), their solubility

286   determind using the ZJU, OUC and NIO protocols was well correlated with that determined

287   using the GIG protocol, and the slopes obtained from correlation analysis, which ranged from

288   0.98 to 1.09 (Table 3), were all close to 1.

289   Since As, Cr, Pb and Zn were not measured using the NIO protocol, we only compared

290   GIG results with ZJU and OUC results for these four elements. As shown in Figures S3, S4,

291   S6 and S8, the solubility of As, Cr, Pb and Zn determined using ZJU and OUC protocols was

292   well correlated with that determined using the GIG protocol, and the slopes (0.87-1.01, as

293   summarized in Table 3) were close to 1.
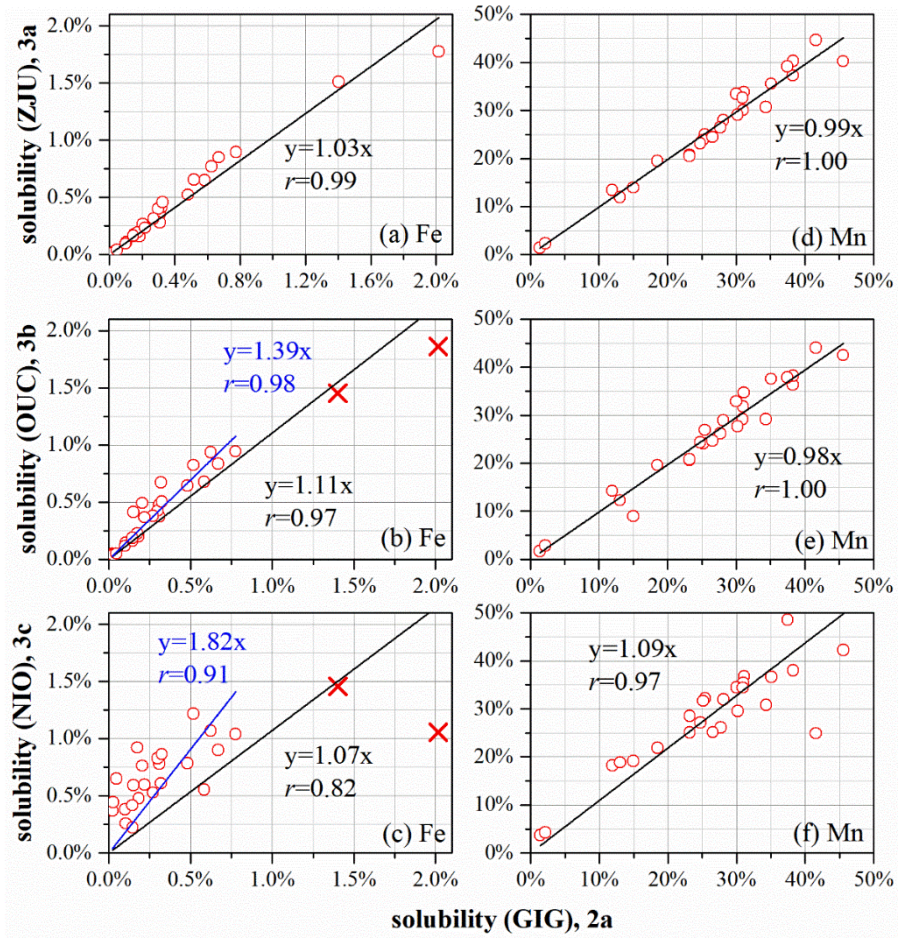
Figure 4. Solubility of Fe and Mn determined using the GIG protocol vs. those determined using ZJU, OUC and NIO protocols: (a) Fe, GIG vs. ZJU; (b) Fe, GIG vs OUC; (c) Fe, GIG vs. NIO; (d) Mn, GIG vs. ZJU; (e) Mn, GIG vs OUC; (f) Mn, GIG vs. NIO. Black lines represent fitting when all the data points are included, and blue lines represent fitting when outliers (represented by red crosses) are excluded.

To summarize, although the four ultrapure water batch leaching protocols differ in agitation method, contact time and/or filter pore size (GIG: shaking, 2 h contact time, 0.22 μm filter pore size; ZJU: sonication, 1 h contact time, 0.22 μm filter pore size; OUC: sonication, 1 h contact time, 0.45 μm filter pore size; NIO: sonication, 1 h contact time, 0.22 μm filter pore size), for the nine elements examined in this intercomparison study, their solubility determined

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306   using the four protocols in general showed good agreement. This is consistant with the results

307   presented in Sections 3.1-3.3, where we found that the effects of agitation method (shaking vs.

308   sonication), contact time (1 vs. 2 h) and filter pore size (0.22 vs. 0.45 μm) were rather limited.

309   The solubility of Fe and Al determined using the NIO protocol deviated considerably from

310   those determined using the GIG protocol, probably because Fe and Al solubility was very low

311   (mostly <2%) and small change in leaching protocol may cause significant change in the

312   amounts of Fe and Al dissolved.

4 Conclusion

314   Ultrapure water batch leaching is widely used in atmospheric research to determine

315   aerosol trace element solubility, and the specific leaching protocols used in different labs can

316   still vary in agitation methods, contact time, and filter pore size. It is yet unclear to which extent

317   the difference in these experimental parameters would affect the reported aerosol trace element

318   solubility; in other words, it remains to be examined whether solubility reported by previous

319   studies which used different ultrapure water batch leaching protocols is comparable.

320   We examined the effects of agitation methods, filter pore size and contact time on the

321   reported solubility of nine aerosol trace elements, including Fe, Al, As, Cr, Cu, Mn, Pb, V and

322   Zn. It was found that the difference in agitation methods (shaking vs. sonication), filter pore

323   size (0.22 vs. 0.45 μm), and contact time (1 vs. 2 h) only led to small and sometimes

324   insignificant difference in the reported solubility. We further compared aerosol trace element

325   solubility determined using four widely used ultrapure water leaching protocols which differ

326   in agitation methods, filter pore size and/or contact time, and in general the solubility

327   determined using the four protocols was found to be in good agreement. Therefore, aerosol

328   trace element solubility determined in previous studies using ultrapure water batch leaching

329   may be comparable. Trace elements were analyzed using similar methods (ICP-MS) in our

330   present work and thus essentially we only examined the effects of leaching protocols;

331   nevertheless, other methods were also used by some previous studies to measure trace elements

332   (Fang et al., 2015; Zhu et al., 2022), probably causing additional uncertainties.

333   Aerosol trace element solubility is an operationally defined term (Baker and Croot, 2010;

334   Meskhidze et al., 2019), and strongly depends on the leaching protocol employed. A number

335   of leaching protocols have been used in previous studies to extract dissolved trace elements,

336   making it very challenging to compare solubility reported in different studies (Perron et al.,

337   2024). In order to reduce uncertainties in aerosol trace element solubility, it is necessary to

338   formulate standard operating procedures for frequently-used aerosol leaching protocols. Our

339   current work suggests that although ultrapure water batch leaching protocols used by different

340   labs vary in specific experimental parameters, the determined aerosol trace element solubility

341   showed good agreement; furthermore, ultrapure water batch leaching is operationally simple

342   and does not introduce any other chemical species which may interfer analysis of water-soluble

343   inorganic ions and organics. Therefore, we recommend ultrapure water batch leaching to be

344   one of the reference leaching schemes. We note that large difference in solubility determined

345   using the four common leaching protocol we examined was also observed for Fe and Al (Table

346   1); moreover, the experimental parameters examined in this work do not cover the whole ranges

347   of these used by various ultrapure water batch leaching protocols used in previous studies. As

348   a result, before a standard operating procedure can be formulated for ultrapure water batch

349   leaching, the community will need to reach consensus on agitation methods, contact time and

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350 filter pore size, and further intercomparison studies, preferentially with more labs involved,

351 will be very helpful.

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353 **Data availability.**

354 Data are available upon request (Mingjin Tang: mingjintang@gig.ac.cn).

355 **Author contributions.**

356 Rui Li: methodology, formal analysis, investigation, writing - original draft, writing - review

357 & editing; Prema Piyusha Panda: formal analysis, investigation; Yizhu Chen: investigation;

358 Zhenming Zhu: investigation; Fu Wang: investigation, supervision; Yujiao Zhu: resources; He

359 Meng: resources; Yan Ren: resources, supervision; Ashiwini Kumar: resources, writing -

360 review & editing, supervision; Mingjin Tang: conceptualization, methodology, resources,

361 writing - original draft, writing - review & editing, supervision.

362 **Competing interests.**

363 Mingjin Tang is a member of the editorial board of Atmospheric Measurement Techniques.

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