Maagi *et al. Int J Concr Struct Mater (2019) 13:62*
https://doi.org/10.1186/s40069-019-0371-y

International Journal of Concrete Structures and Materials

RESEARCH

Open Access

Effect of Nano-SiO_2, Nano-TiO_2 and Nano-Al_2O_3 Addition on Fluid Loss in Oil-Well Cement Slurry

Mtaki Thomas Maagi, Samwel Daud Lupyana and Jun Gu*

Abstract

In this article, incorporation of nano-SiO_2 (NS), nano-TiO_2 (NT) and nano-Al_2O_3 (NA) particles and their binary and ternary blends on water filtration in oil-well cement slurry was examined. The nanoparticle contents were chosen at proportions corresponding to 1, 2, 3 and 4% based on the weight of cement. The experimental specimens were tested at three various temperatures of 70, 80 and 90 °C using a gas pressure of 1000 psi. The quantity of water filtrate collected was measured in milliliter (mL) at 30 min after the test begins. The results consistently indicate that an additional of NS, NT and NA particles independently, reduced the loss of liquid in cement, and its performance varies with temperature and the nanoparticle dosages. The 3% NS usage delivered strong evidence in lessening fluid loss compared to the other results by reducing the loss up to 72%. When collective impact of nanomaterials was determined, the fluid retainment was also improved. The replacement of 4% NST reduced fluid loss by the most compared to NSA and NTA binary groups. In-case of ternary combination, NSTA showed a highest reduction of the water loss by 58–60% likened to the plain samples at the concentration of 4%. The key investigation of this paper clearly suggested that the efficacy of Class G cement having nanoparticles to trap its water is dependent on the nanoparticle contents. The lessening of water filtration might be explicated by the filling capability of nano-scale particles. Nanoparticles can plug the openings within the oil-well cement slurry matrix, thus promoting the retainability of water. Besides, nanoparticles quickens the hydration products by creating dense interlocking C–S–H gels for bridging cement grains and forming enclosed structure which can stop the liquid from escaping the slurry.

Keywords: oil-well cement, fluid loss, nanoparticles, nano-SiO_2, nano-TiO_2, nano-Al_2O_3

1 Highlights

- Designing of cement slurry without filtration control agent causes much water to be lost from the slurry, thereby affecting other properties of the slurry such as viscosity and thickening time.
- Nanoparticles help control fluid loss from the cement slurry, and its performance varies with temperature, type and the content of nanoparticles.
- Nanoparticles fill the voids between cement grains, resulting in the immobilization of free water.

2 Introduction

Oil-well cementing refers to a process of pumping cement in the annular space to bond the steel casing strings to the borehole surface (Newlove et al. 1984; Nelson and Guillot 2006). The crucial purposes of the cementing process are sealing (prevention of fluid communication between different zones), provision of structural sustenance for the casings and sheltering the casings from attack by corrosive fluids (Fakoya and Shah 2017; Newlove et al. 1984). One of the challenging problem during primary oil-well cementing is an out-flow of fluid from the pumped cement to the permeable formation. This loss of liquid is disadvantageous

*Correspondence: gujun@cug.edu.cn
Department of Petroleum Engineering, Faculty of Earth Resources, China University of Geosciences, Wuhan 430074, China
Journal information: ISSN 1976-0485 / eISSN 2234-1315

since since it can cause de-hydration of the slurry before its normal setting time (Newlove et al. 1984). When this occurs it prevents the pumped slurry to reach its final position and leaving unwanted cement slurry in the casing becoming one of the primary reasons for unsuccessful cementing. In order to accomplish effectual placement of the slurry as it is pumped to plug the annulus, preclusion of filtration loss is important. To control the liquid loss from the cement, it is necessary to lessen the oil-well cement permeability and trap water during the initial cement setting process, effectively preventing speedy drying up of the designed cement slurry (Brandl et al. 2011). This helps to mitigate solids settling by enhancing fluid stability.

According to the work of (Nelson and Guillot 2006), a well that lacks a worthy cement job may never attain its full production potential. In addition, they advocated that oil-well cement need to be adjusted to enhance its chief functions. In that regard, various authors recently have shown intense concentration on using nanoparticles to boost the useful lifecycle of the oil-well and cut outlays related to the reparation or damages due to failed cement sheath. Nano-SiO_2 (NS), nano-TiO_2 (NT) and nano-Al_2O_3 (NA) have exposed excessive potential in up-lifting the performance of oil-well cements (Jayapalan et al. 2010; Mohseni et al. 2015; Pang et al. 2009; Patil and Deshpande 2012; Quercia and Brouwers 2014; Santra et al. 2012; Senff et al. 2012). Due to their nanoscale dimension, nanoparticles possess exceptional prominent characteristics that differ basically with their macro-sized counterparts. Apart from their size effect, nanoparticles have special characteristics including enormous surface areas per volume ratios and higher chemical reactivity which triggers their ability to modify properties in cement (Amanullah and Al-Tahini 2009; Campillo et al. 2007; El-Diasty and Ragab 2013; Fakoya and Shah 2017; Patil and Deshpande 2012; Silvestre 2015). Fluid loss is among the cement behavior that can be adjusted by nanoparticles although it has been generally reported by few researchers compared to other properties such as mechanical strength, microstructures and cement hydration. It has been established that when nano-scale powders are used as filtration control agents can effectively diminish fluid loss, improve mechanical behavior of the cement and resist gas migration (Roddy and Duncan 2008). Moreover, (Patil and Deshpande 2012) investigated the capability of nano-SiO_2 on compressive strength, thickening time and fluid loss in oil-well cement. The study disclosed that nanosilica can increase both early and final compressive strength, prevents fluid loss and slacked the thickening time of cement. Likewise, Ershadi et al., revealed that nanosilica promotes compressive strength while reducing fluid loss and thickening time (Ershadi et al. 2011).

More attentions are paid on the increasing latest studies on nano-scale particles to modify properties and performance of oil-well cement, and the outcomes acquired from such probes. However, most of the previous studies associated with the application of nano- sized materials in oil-well cementing are focused on a single nanoparticle type. Researches on the effects of SiO_2, TiO_2 and Al_2O_3 nanoparticles to enhance properties of oil-well cement in a combination of two or more kinds of nanoparticles are lacking. The present study examines the effects of nano-SiO_2, nano-TiO_2 and nano-Al_2O_3 particles and their binary and ternary blends on fluid loss in oil-well cement slurry. It is plausible that findings from this probe can provide evidences for the usefulness of nano-scale materials in oil-well cement especially in the case of combined nanoparticles.

3 Experimental Programs

3.1 Experimental Materials

The experimental materials include Class G oil-well cement, corresponded to American Petroleum Institute (API) Specification 10A, (produced by Jiahua Enterprises Corp., Sichuan China, its clinker compositions are concisely accessible in Table 1) and nano-SiO_2, nano-TiO_2 and nano-Al_2O_3 particles (purchased from Guangzhou Probig Fine Chemical Co., Ltd, China, its technical specifications are provided in Table 2).

Table 2 Properties of nanoparticles

Species	Diameter (nm)	Color	Surface volume ratio (m^2/g)	pH value	Purity (%)
SiO_2	20	White	220	5.7	99.9
TiO_2	20	White	160	6.8	99.9
Al_2O_3	20–30	White	50–80	6.8	99.9

Table 1 Chemical and mineral composition of Class G oil-well cement used

Chemical composition (wt%)					Mineralogical composition (wt%)				
SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	C_3S	C_2S	C_4AF	C_3A
23.056	2.86	3.52	65.2	1.79	2.12	59.890	16.756	10.70	1.63

3.2 Experimental Methods

3.2.1 Slurry Design and Fluid Loss Test

The oil-well cement blending and its fluid loss tests were done according to API standards (API 2013). The TG-71 fluid loss apparatus was used to quantify the rate of filtration of the designed cement slurry (Fig. 1). The fluid loss instrument works by simulating the down-hole conditions where fluid loss can occur, when cement is injected to plug the annulus. The nano-sized particles of 1, 2, 3 and 4% by weight of cement were thoroughly mixed with the Class G Oil-well cement and water at 0.45, water-to-cement ratio. Then, the cement was methodically blended using a high speed blender (4000 r/min for 15 s, then 12,000 r/min for 35 s according to AP1 10A standards) (API 2010). The cement slurries contained in a test cell were placed into the warming jacket to test the loss of fluid under different temperatures of 70, 80 and 90 °C at 1000 psi gas pressure. The liquid filtrate collected was measured in milliliter (mL) at 30 s, and 1, 5, 10, 15, 20 and 30 min after the test begins. All the filtrate left the cement through the sieves in less than 30 min, thus the API Fluid loss was calculated using the formula indicated in Eq. 1.

$$Calculated\ API\ fluid\ loss = 2V_t\left(\frac{5.477}{\sqrt{t}}\right) \quad (1)$$

where Vt stands for volume of filtrate (mL) collected at time t (min).

Fig. 1 TG-71 Fluid loss tester

4 Experimental Results

4.1 The Effect of Adding Single Nanoparticles

The liquid loss from the oil-well cement with varying contents of nanoparticles was tested, and the temperature range was between 70 and 90 °C. Figure 2 a–c shows the fluid loss performance results obtained for various specimens with single nanoparticles. As the Figure indicates, the loss of fluid from the slurries decreased gradually with the accruing dosage of the nanoparticles. The highest rate of the cement to trap its water was obtained when nanoparticles of 3% NS concentration was employed. In general, at all observed temperatures, the nanoparticles showed useful results, and the filtration loss of the slurry was below 100 mL which is generally considered to be adequate for most primary cementing operations. Compared to the control samples, the slurry containing NS particles reduced the loss of fluid by 70–72%, while the specimens containing NT and NA powders reduced the loss by 61–63% and 57–60% respectively. However, the samples containing 4% NS thickened instantly after heating up the slurry to the required temperatures, thus that test was not carried out. One interesting observation is that NS makes cement slurry thicker and quickens the cement hydration (Silvestre 2015). When NS are added into cement, it shows a straight effect on the ratio of water needed in the blend. Such kind of behavior clearly approves that incorporation of nano-scale particles into Portland cement requires more water to retain the pumpability of the slurry. If the proportion of water is retained, an additional of the NS dosage will stimulate the filler effect, plugging the openings between cement particles and hence preventing the flexibility of water. Numerous literatures have clearly reported that the optimal dosages of the nano-sized particles in cement pastes are influenced by the average particle size (Quercia and Brouwers 2014). Thus, due to its huge specific surface area, NS showed high effect on slurry fluidity indicating that the cement replacement of 4% was overuse of NS.

4.2 The Effect of Adding Binary Nanoparticles

The results on fluid loss of the cement containing a binary combination of nanoparticles are clearly presented in Fig. 3a–c. As the Figure shows, all the binary samples demonstrated a trend of reducing the filtration loss from the slurry. It can be observed that, the NST samples exhibited the ultimate control of liquid passage overall among the samples at the proportion of 3 and 4%. The slurry specimen containing NST particles reduced the filtration loss by 64–66% compared to the plain samples. This indicates that the efficiency of the slurry to trap its liquid phase is influenced by the extent of the filtration control agents. Overall, nanoparticles

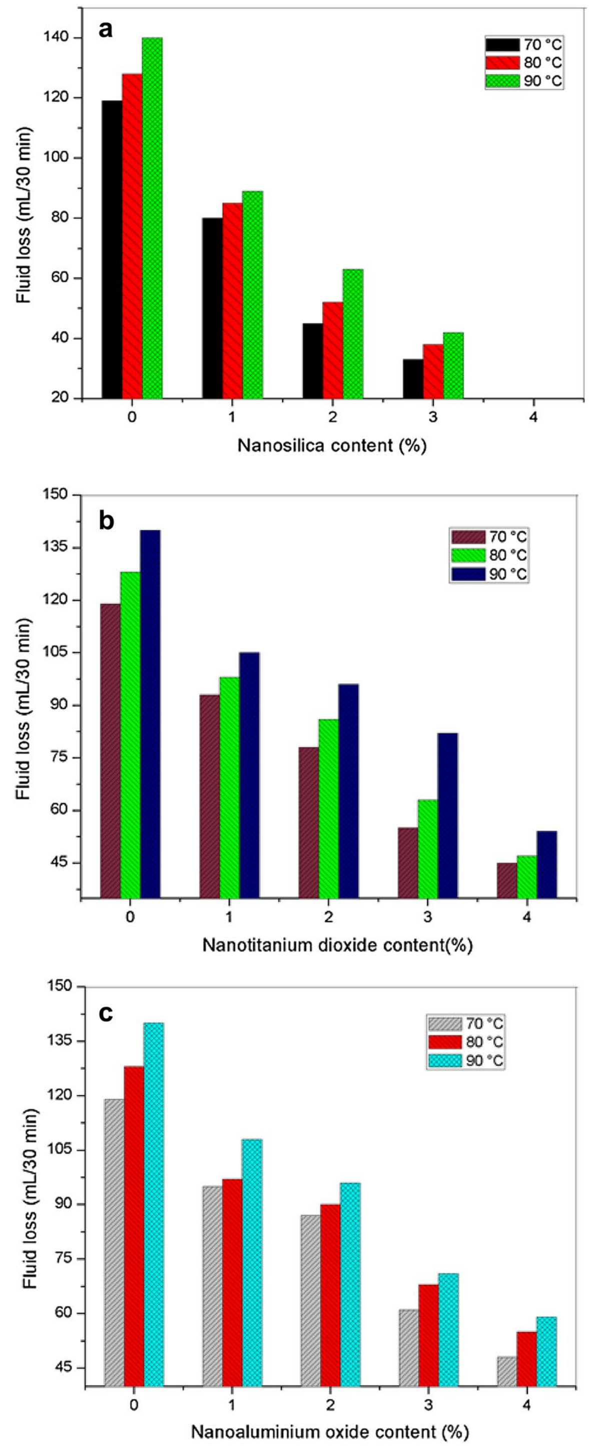

Fig. 2 **a** The effect of adding NS on fluid loss. **b** The effect of adding NT on fluid loss. **c** The effect of adding NA on fluid loss

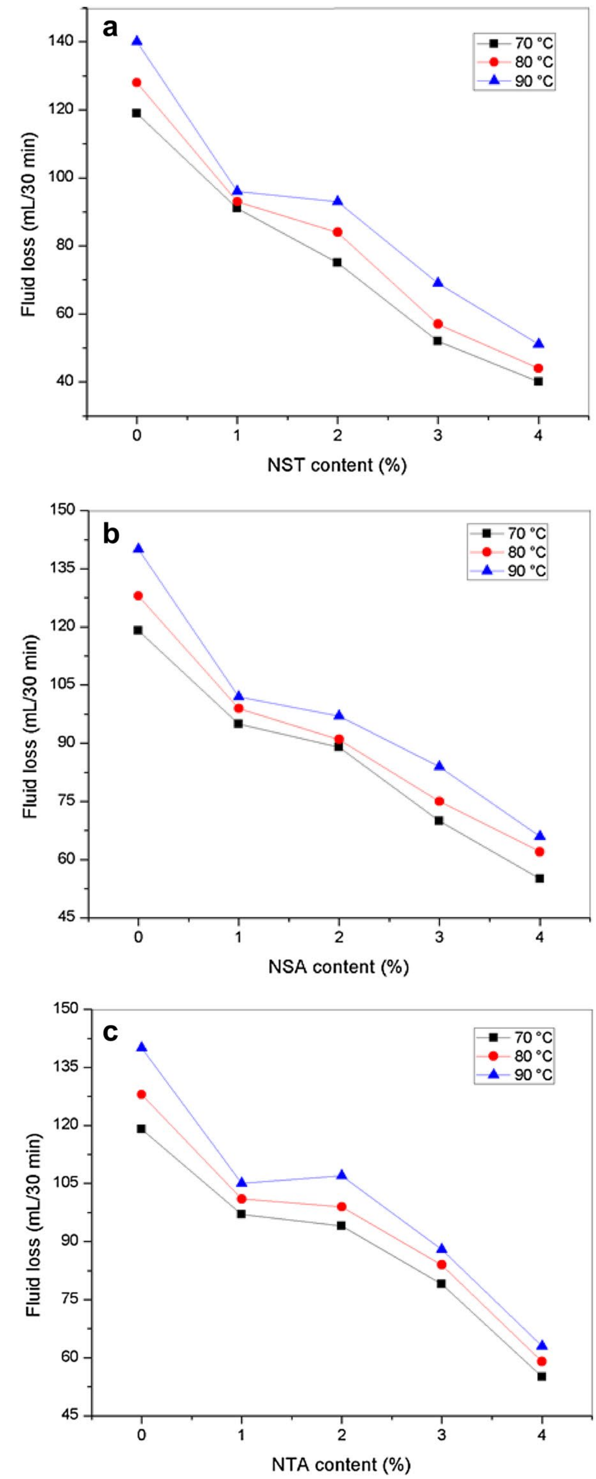

Fig. 3 **a**The effect of adding NST on fluid loss. **b** The effect of adding NSA on fluid loss. **c** The effect of adding NTA on fluid loss

can fill the openings between the cement grains, thus restricting an out-flow of liquid (Ershadi et al. 2011; Roddy and Duncan 2008). The slowdown of filtration loss could be explained by the filler effect displayed by nanoparticles and its aptitude to form a more intense cementitious structure. Another significant finding specified that an additional of binary powders in the

Table 3 The effect of adding ternary nanoparticles on fluid loss control

Batch of slurry	Nanoparticle dosage (%)	Calculated API fluid loss (mL/30 min)		
		70 °C	80 °C	90 °C
Control	0	119	128	140
NSTA1	1	92	96	99
NSTA2	2	83	89	93
NSTA3	3	61	63	74
NSTA4	4	48	52	59

blend of NSA and NTA reduced the filtration loss by 52–54% and 54–55% respectively. Thus, blending of binary nanoparticles into oil-well cement slurry could have a significant potential for controlling fluid loss.

4.3 The Effect of Adding Ternary Nanoparticles

For the cement slurry prepared by adding ternary nanoparticles, its results are displayed in Table 3. As briefly displayed in Table 3, ternary combination of nanoparticles (NSTA) showed effective properties as a fluid loss reducing material compared to the control specimens. It is clearly observed that there is a trend of decreasing fluid loss depending on the ratio of nano-scale particles and temperature variations. The out-flow of liquid decreased gradually with the cumulative nanoparticles content. Conversely, the filtration loss was noticeably found to increase progressively as temperature was adjusted to 90 °C. However, the samples with ternary combination of nanoparticles showed an effective performance on restricting fluid loss. In general, the fluid collected in all ternary samples was less than 100 mL. Thus, similar to binary combination of nanoparticles, blending of ternary nanoparticles into oil-well cement slurry could also have a significant potential for controlling fluid loss. It seems that, the nano-scale particles were well able to place between the empty pore of cement matrix and did not let the solid part of the slurry to be separated from the liquid, thus stopping the escapement of water from the cement matrix (Ershadi et al. 2011). This phenomenon helps to diminish the loss of fluids from the cement slurry.

5 Conclusion

A number of tests were successfully conducted to examine the helpfulness of NS, NT and NA nanoparticles and their combined effects on fluid loss in oil-well cement. Based on the investigational outcome, the following are our conclusions;

1. Designing of conventional cement slurries without any additional filtration control agent causes much water to be lost from the set-cement, thereby affecting other characteristics of the slurry such as viscosity, thickening time and compressive strength.
2. Additional of NS, NT and NA nanoparticles control the out-flow of fluid from the slurry, and its performance varies with temperature and the nanoparticles dosages. The ratio of 3% NS reduced the filtration loss more significantly compared to other single nanoparticles. However, excessive amount of NS decreases the slurry fluidity.
3. By comparison regarding the combined of effect of nanoparticles, the binary combination mixtures containing NST demonstrated the superlative results likened to other binary groups followed by ternary combination of NSTA samples. The highest rate of the blended cement to trap its liquid phase was found at a dosage of 4% nanoparticles. This indicates that the efficiency of the oil-well cement slurry containing nanoparticles to retain water depends intensely on the concentration of the nanoparticles.
4. In summary, by relating all the examined samples containing nanoparticles, NS provided the best performance in controlling fluid loss. The optimal ratio of NS in the blended cement system was 3%.

Supplementary information

Supplementary information accompanies this paper at https://doi.org/10.1186/s40069-019-0371-y.

Additional file 1: Experimental raw data for fluid loss test.

Acknowledgements
This investigation was sponsored by Grant Nos: 51774258 and 41972326 from the National Natural Science Foundation (China). The probe was also backed by the Grant No: 2017ZX05009003-003 under the National Science and Technology Major Project (China). Additionally, the authors extremely thank China University of Geosciences (Wuhan) for the support under the Grant No: CUGQYZX1710.

Authors' contributions
MM conceived and planned the experiment. MM and SL performed the experiment, analyzed data and prepared the manuscript. JG controlled the research. All the three authors discussed the results and produced the last version of the manuscript. All authors read and approved the final manuscript.

Availability of data and materials
The datasets used to defend the findings of this experimental work are incorporated in the article.

Competing interests
The researchers proclaim that no contending interests between them.

Received: 21 March 2019 Accepted: 21 September 2019
Published online: 25 November 2019

References

Amanullah, M., & Al-Tahini, A. M. (2009). Nano-technology-its significance in smart fluid development for oil and gas field application. In A. M. Al-Tahini (Ed.), *SPE Saudi Arabia Section Technical Symposium*. AlKhobar, Saudi Arabia: Society of Petroleum Engineers.

API 10A (2010) Specification for cements and materials for well-cementing. Amer Petr Inst.

API 10B-2 (2013) Recommended practice for testing well-cements. Amer Petr Inst.

Brandl, A., Cutler, J., Seholm, A., et al. (2011). Cementing solution for corrosive well environment. *SPE Drill & Compl, 2,* 208–219.

Campillo, I., Guerrero, A., Dolado, J., et al. (2007). Improve of initial mechanical strength by nano-alumina in belite cement. *Materials Letters, 61*(8), 1889–1892.

El-Diasty, A. I., & Ragab, A. M. S. (2013). Applications of nanotechnology in the oil & gas industry: Latest trends worldwide & future challenges in Egypt. In *North Africa Technical Conference and Exhibition*. Society of Petroleum Engineers.

Ershadi, V., Ebadi, T., Rabani, A. R., et al. (2011). The effect of nano-SiO_2 on cement matrix permeability in oil-well to decrease the pollution of receptive environment. *Journal of Environmental Science and Development, 2,* 128–132.

Fakoya, M. F., & Shah, S. N. (2017). Emergence of nano-technology in the oil & gas industry: Emphasis on the application of silica nano-particles. *Petrol, 3*(4), 391–405.

Jayapalan, A., Lee, B. Y., Fredrich, S. M., et al. (2010). Influence of addition of anatase TiO_2 nano-particles on early age properties of cement-based materials. *Transportation Research Record, 2141*(1), 41–46.

Mohseni, E., Miyandehi, B. M., Yang, J., et al. (2015). Single and combined effects of nano-SiO_2, nano-Al_2O_3 and nano-TiO_2 on the mechanical, rheological and durability properties of self-compacting mortar containing fly-ash. *Construction and Building Materials, 84,* 331–340.

Nelson EB, Guillot D (2006) Well Cementation. Developments in Petroleum Science Book Series, Schlumberger

Newlove JC, Portnoy RC, Schulz, DN et al (1984) Fluid loss control in oilfield cements. USA Patent No. 4,480, 693

Pang, X., Boul, P. J., & Jimenez, W. (2009). Nano-silica as accelerators in oil-well cementing at low temperature. *SPE Drilling & Completion, 29*(01), 98–105.

Patil, R. C., & Deshpande, A. (2012). Use of nanomaterials in cementing applications. In *SPE International Oilfield Nanotechnology Conference and Exhibition*. Society of Petroleum Engineers, Noordwijk, The Netherlands

Quercia, G., & Brouwers, H. J. H. (2010). Application of nano-silica (nS) in concrete mixtures. In *8th fib PhD Symposium in Kgs*.

Roddy, C. W., Chatterji, J., & Cromwell, R. (2009). Well treatment compositions and method utilizing nano-particles. USA, Patent no, 02777116 AI U.

Santra, A. K., Boul, P., & Pang, X. (2012). Influence of nanomaterials in oilwell cement hydration and mechanical properties. In *SPE International Oilfield Nanotechnology Conference and Exhibition*. Society of Petroleum Engineers, Noordwijk, The Netherlands.

Senff, L., Hotza, D., Lucas, A., et al. (2012). Effect of nano-SiO_2 and nano-TiO_2 on the rheological behavior and the hardened properties of cement-mortars. *Mat Sci Eng, 532,* 354–361.

Silvestre, J. P. T. (2015) Nanotechnology in construction: Towards structural applications. Civil Eng Master Thesis Report, Lisbon Portugal.

Publisher's Note

Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.