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TRANSFER LEARNING FOR NEURAL NETWORK SOLUTION OF AN INVERSE PROBLEM IN OPTICAL SPECTROSCOPY*

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Inverse problem of spectroscopy

Inverse problems are the type of problems
when the system parameters are determined
from the observed data describing the system state.

The inverse problem of spectroscopy is the determination of characteristics of an object by its spectrum.

The present study considered the problem
of determining the concentrations of heavy metal ions in water
by Raman, infrared and optical absorption spectroscopy.

Actuality

The problem of determination of concentrations of substances dissolved in water is very important for:

- Oceanology
- Ecological monitoring
- Control of industrial and waste waters

This problem is required to be solved in non-contact express mode with acceptable precision.

Actuality

Traditional chemical methods:

- provide a high enough accuracy of determining the concentration of ions
- contact
- each test requires substantial time
- each test is individual for each ion
- need for laboratory equipment and special reagents
- requires sufficient staff qualification

Therefore, spectroscopic methods are considered as an alternative.

Actuality

Raman, infrared and absorption spectroscopy as an alternative:

- express
- non-contact
- The shapes of the spectra of solutions are highly sensitive to changes in its ionic composition and to the concentrations of ions
- At the moment there is no adequate mathematical model capable of describing these changes in the shape of Raman, infrared and absorption spectra.

For processing of such data, it is suggested to use methods of machine learning.

Transfer Learning

- Training neural networks is a computationally expensive procedure requiring large datasets that are not always available
- Neural networks pre-trained on a dataset of some problem may be used to solve other similar problems after adjusting them to a new problem by fine tuning on a relatively small data array
- Such method is called **Transfer Learning**
- The size of the adjustment array may be limited also by the amount of available data

Purpose of the study

- Natural waters are characterized
  by a large amount of dissolved organic matter
- This matter has strong fluorescence
  that is variable by its spectrum and by its intensity
- Its spectrum overlaps with Raman spectrum
  of water and ions of inorganic salts
- It affects optical absorption spectrum of water
- Therefore, NN trained on spectral data of solutions
  made in distilled water, degrade much when applied
  to samples taken or made in river water

The goal of this study is to use transfer learning approach
to adapt such NN to perform well in river water

Problem statement

The problem considered in this study was to identify and determine the concentrations of 8 ions contained in a multi-component solution of 10 salts by their Raman, infrared and optical absorption spectra.

Determined ions:

- Anions: SO_4^{2-}, NO_3^-
- Cations: Zn^{2+}, Cu^{2+}, Li^+, Fe^{3+}, Ni^{2+}, NH_4^+

Salts:

- $Zn(NO_3)_2$, $ZnSO_4$, $Cu(NO_3)_2$, $CuSO_4$, $LiNO_3$, $Fe(NO_3)_3$, $NiSO_4$, $Ni(NO_3)_2$, $(NH_4)_2SO_4$, $NH_4(NO_3)$

The investigated solutions contained 1 to 10 of the salts and 2 to 8 ions. Concentration range of cations – 0-1 M.

Absorption spectroscopy

Experimental setup:

- Shimadzu UV-1800 spectrophotometer

Measurements:

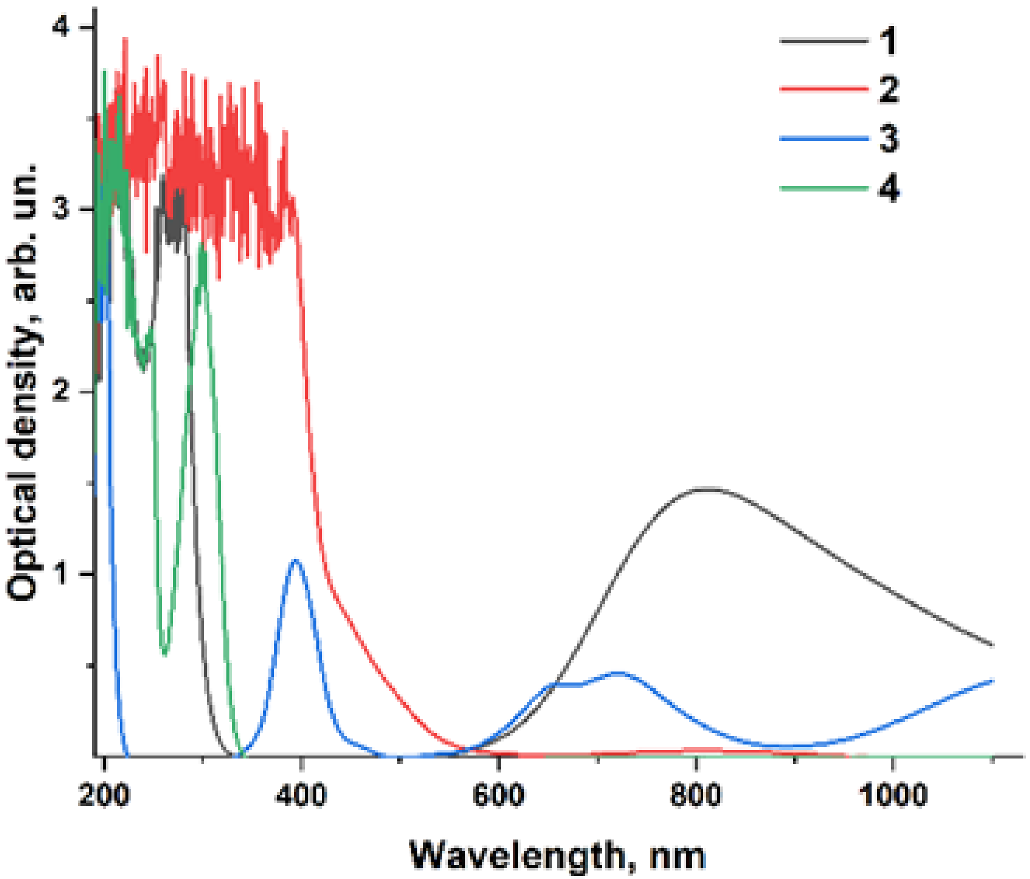
- Measurement range – 190-1100 nm
- Measurement step – 1 nm
- 911 channels

Preprocessing:

- Not performed

Absorption spectroscopy

- Salts: 1 – $CuSO_4$, 2 – $Fe(NO_3)_3$, 3 – $NiSO_4$, 4 – $Zn(NO_3)_2$
- Concentrations of salts – 1 M



Infrared spectroscopy

Experimental setup:

- Bruker Invenio R spectrometer

Measurements:

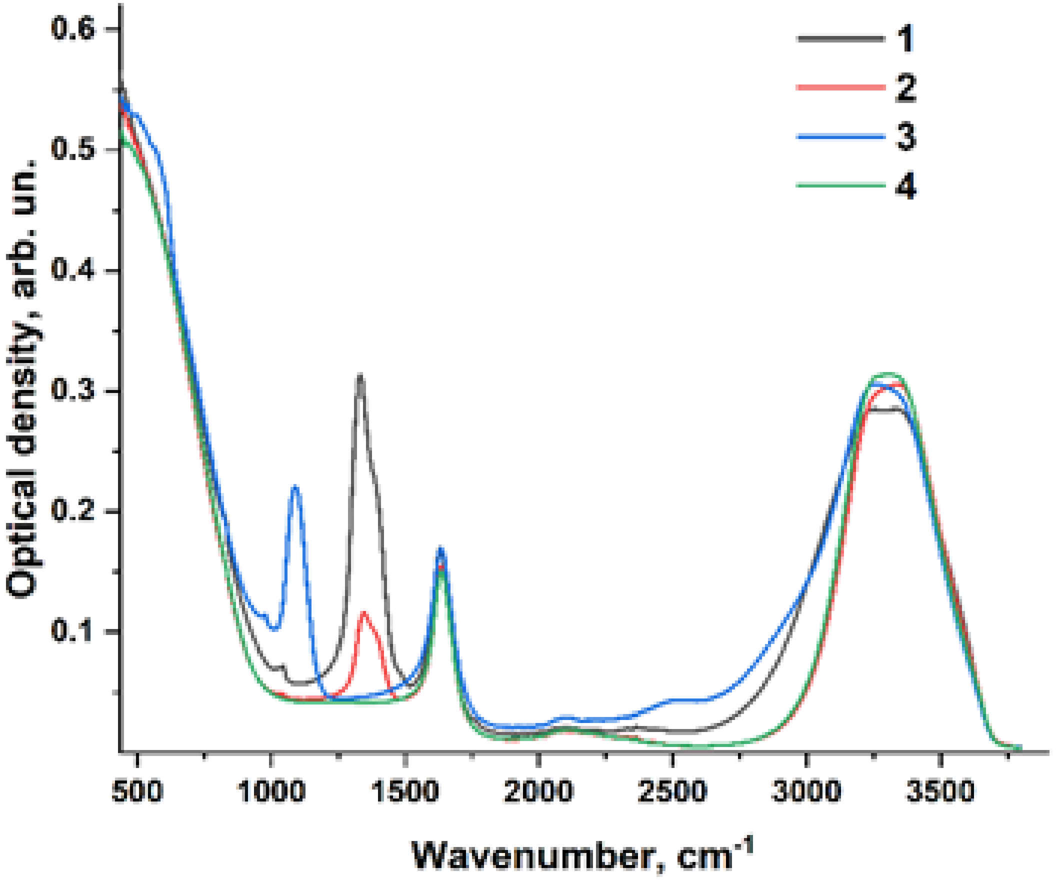
- Measurement range – 400-4500 cm^{-1}
- Resolution – 4 cm^{-1}
- 2126 channels

Preprocessing:

- Not performed

Infrared spectroscopy

- Salts: 1 – $Cu(NO_3)_2$, 2 – $LiNO_3$, 3 – $(NH_4)_2SO_4$, 4 – dist. water
- Concentrations of salts – 1 M



Raman spectroscopy

Experimental setup:

- YAG - laser
  - ✓ Wavelength – 532 nm
  - ✓ Power – 500 mW
- Acton 2500i monochromator
- CCD-camera Horiba Jobin Yvon

Measurements:

- Measurement range – 300-4019 cm^{-1}
- 2598 channels

Preprocessing:

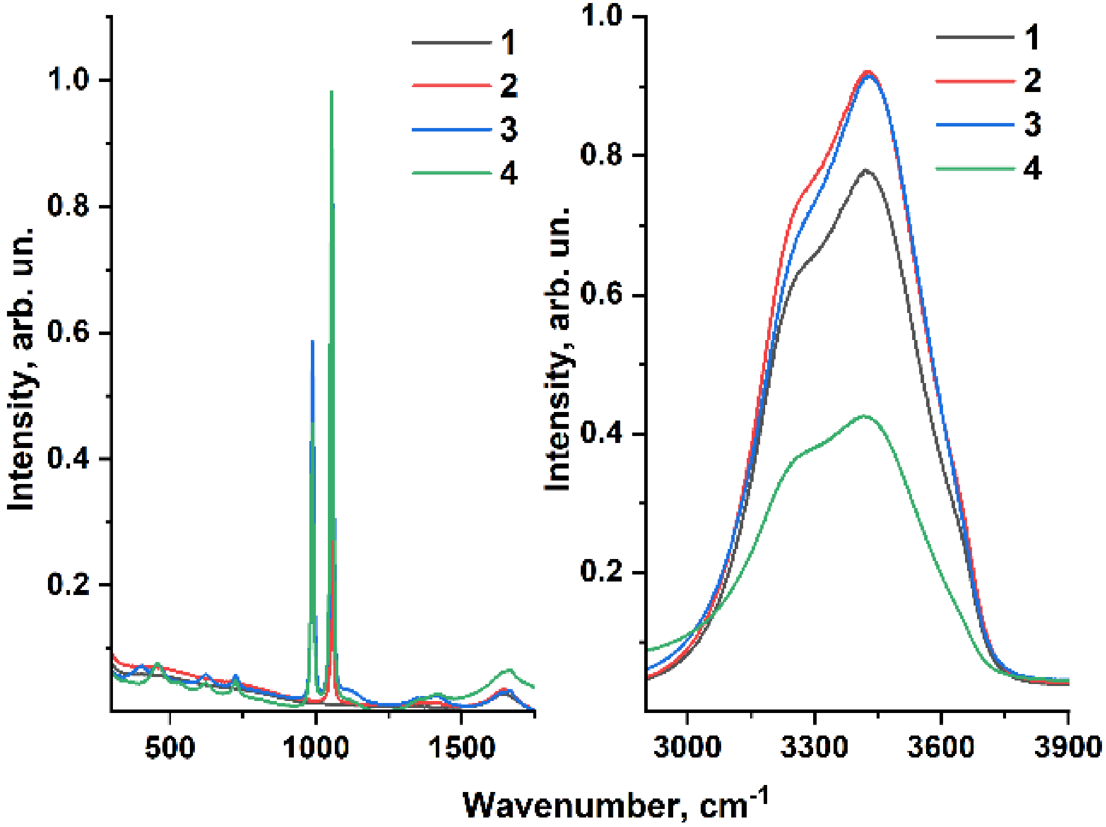
- Subtraction of the pedestal

Raman spectroscopy

- Salts: 1 – dist. water;

  2 – 0.22M $LiNO_3$; 3 – 0.47M $Zn(NO_3)_2$, 0.62M $ZnSO_4$;

  4 – 0.22M $Cu(NO_3)_2$, 0.47M $LiNO_3$, 0.40M $(NH_4)_2SO_4$



Data

Initial dataset:

- Obtained in experiment

Spectra are divided into the following series:

- Basic series – spectra of salts dissolved in distilled water
- “Gold” series – in river water (Moskva river)
- “Silver” series – in river water (rivers Yauza, Bitsa, Setun’)

Number of patterns (samples):

- Basic series – 3760 patterns
- Gold series – 400 patterns
- Silver series – 3 series, 200 patterns each

Data

Split of the basic series:

• Training set	~ 70%	2660 patterns
• Validation set	~ 20%	700 patterns
• Test set	~ 10%	400 patterns

Data dimensionality:

• By input	2 598 + 2 126 + 911	features
• By output	8	parameters

- A separate model was built for each of the determined parameters (autonomous determination)

Neural network parameters

Parameters of NN training:

- Architecture
  - ✓ Perceptron with 3 hidden layers
  - ✓ 64+32+16 neurons in the hidden layers
  - ✓ Activation function: logistic for the hidden layers
    linear for the output layer
  - ✓ Optimization algorithm – SGD
  - ✓ Learning rate for all layers – 0.01
  - ✓ Moment for all layers – 0.9

- Stop training criterion
  - ✓ 500 epochs after minimum of the error on the validation set

Tasks of the study

Program of the computational experiments:

1) Train NN on the distilled water data (basic series), test performance

   Test performance of the trained NN on river water (gold, silver series)

2) Train NN on river water only (gold series)

   Test performance on data of other rivers (silver series)

   Problem: a very small number of training patterns (400)

3) Test **transfer learning** as a solution to this problem:

   NN pre-trained on distilled water data (basic series) –

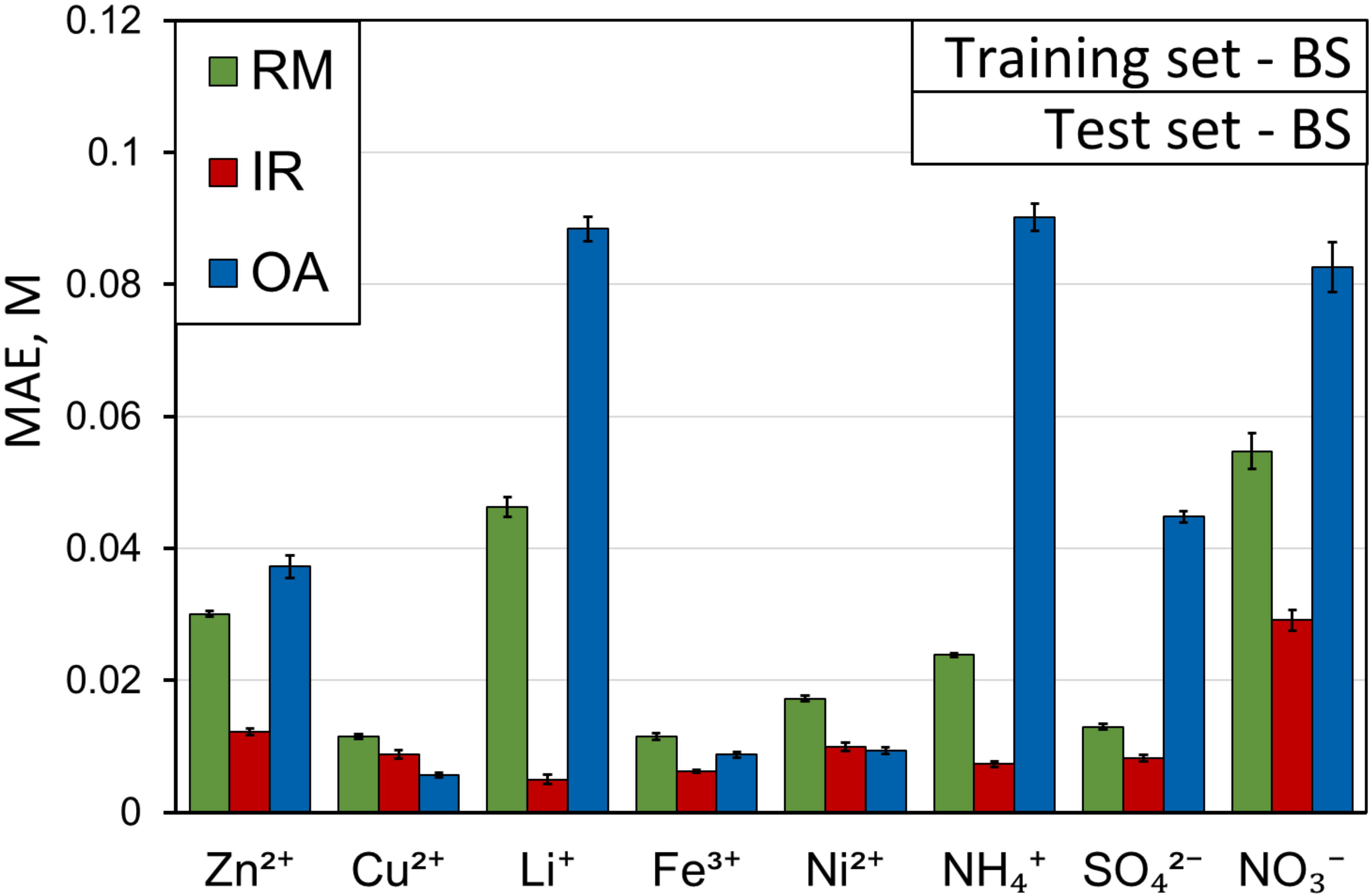
   fine tune on river water data (gold series)

   Test performance on data of other rivers (silver series)

4) Compare the results of all the three approaches

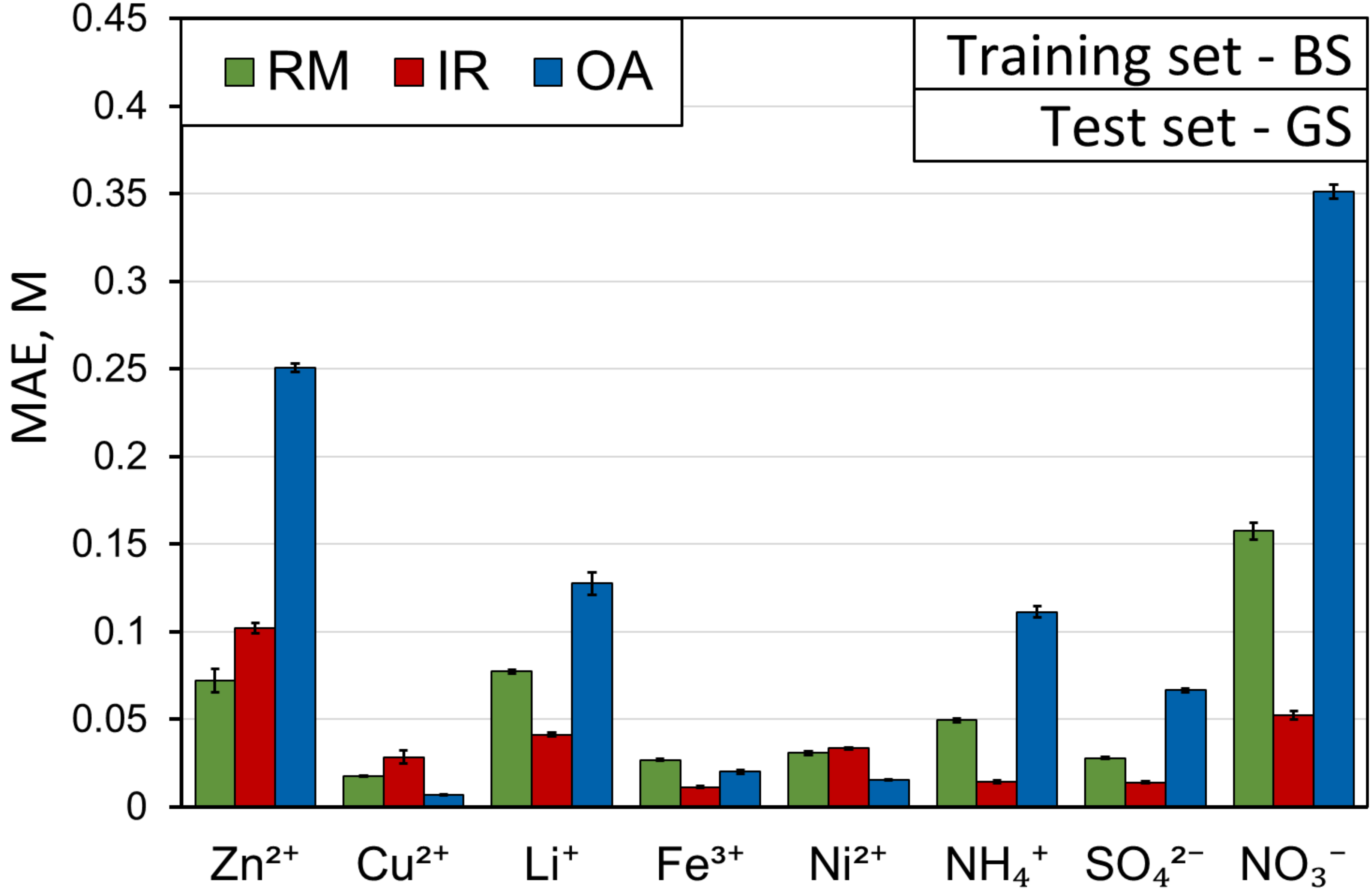
5) Combine with integration of various types of spectroscopy (future)

Results (1)



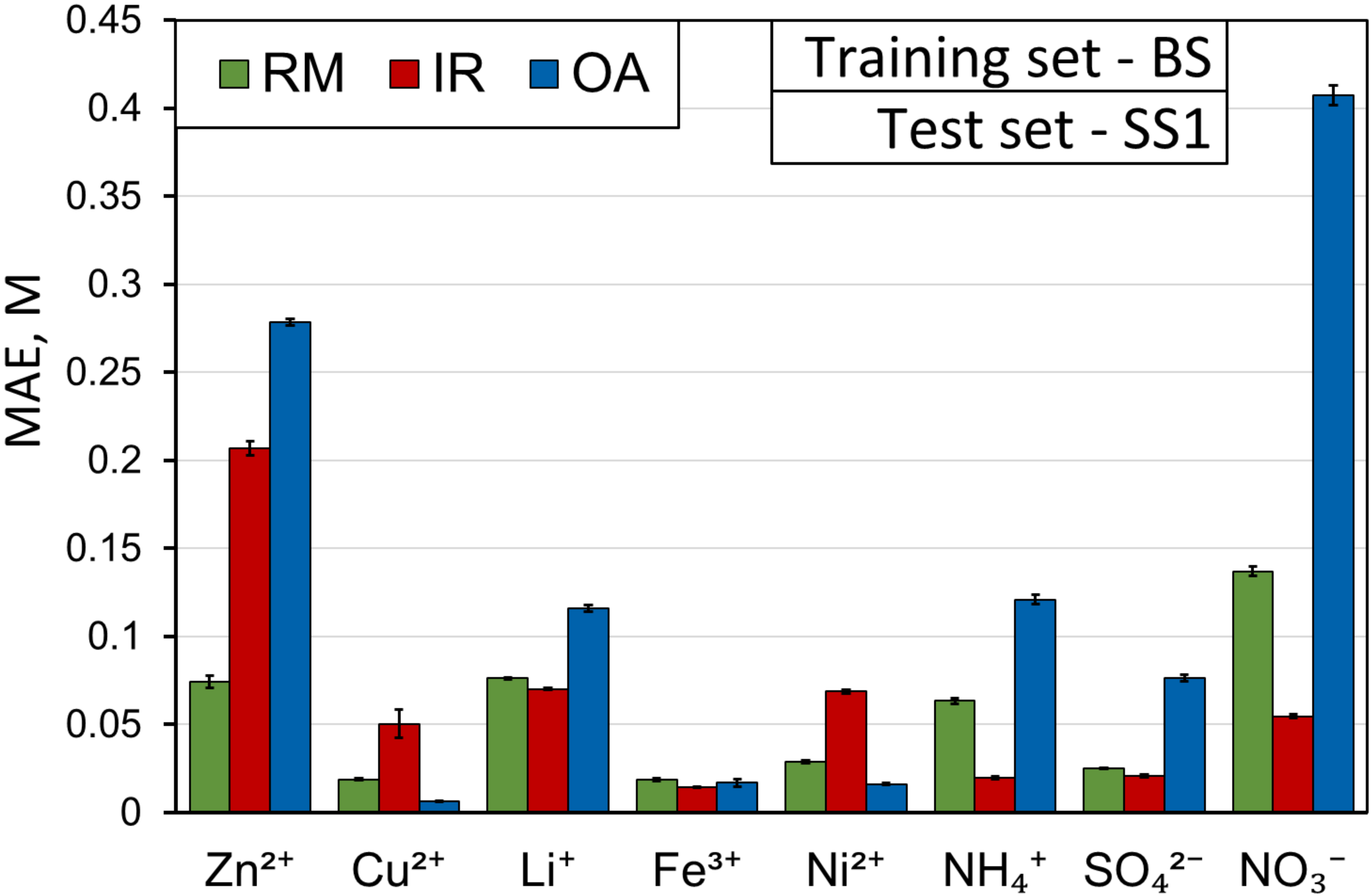
0.12
0.1
0.08
0.06
0.04
0.02
0
MAE, M
RM
IR
OA
Training set - BS
Test set - BS
Zn²⁺
Cu²⁺
Li⁺
Fe³⁺
Ni²⁺
NH₄⁺
SO₄²⁻
NO₃⁻

Results (1)

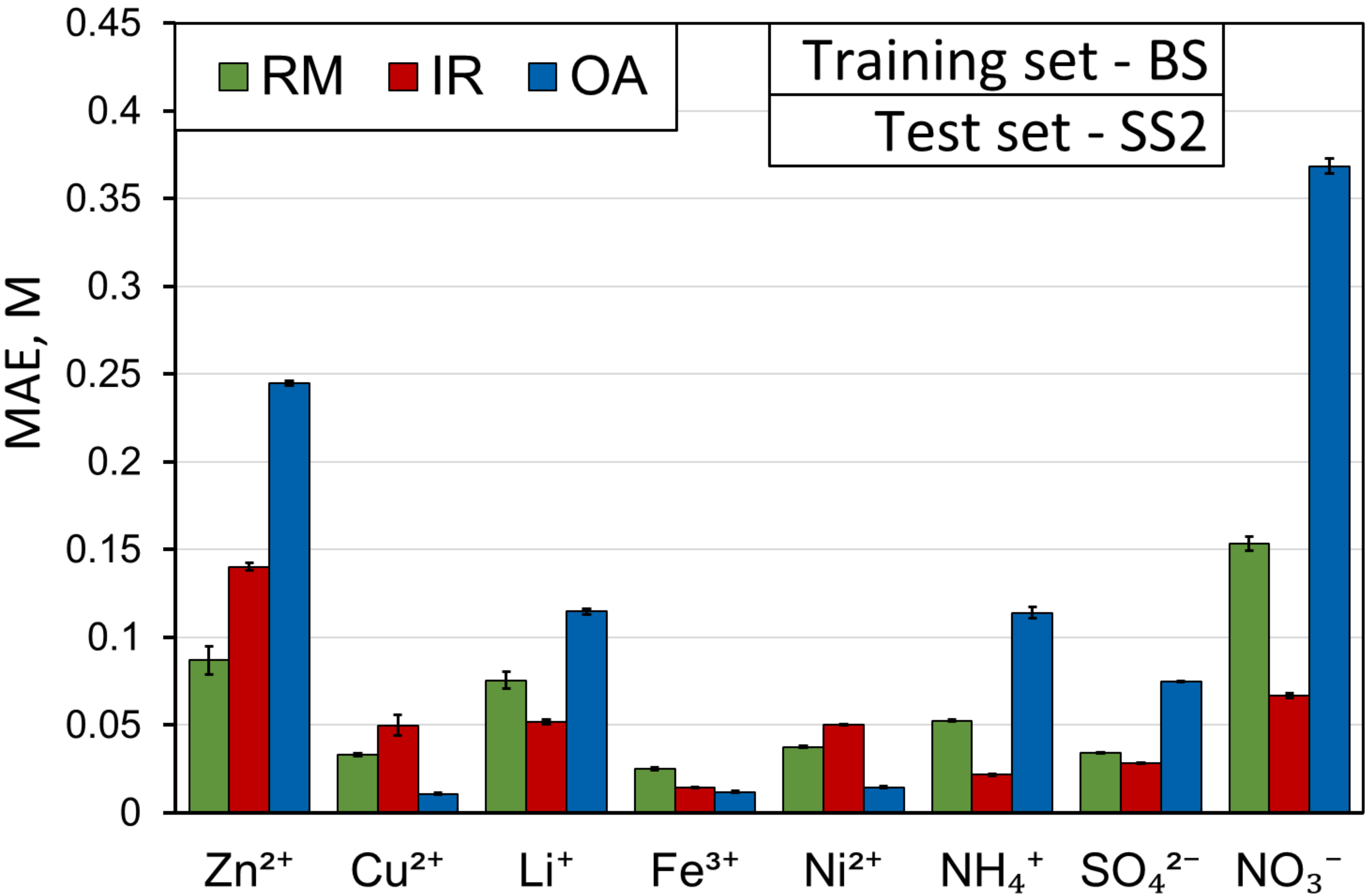


RM
IR
OA
Training set - BS
Test set - GS
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn^{2+}
Cu^{2+}
Li^{+}
Fe^{3+}
Ni^{2+}
NH_4^{+}
SO_4^{2-}
NO_3^{-}

Results (1)

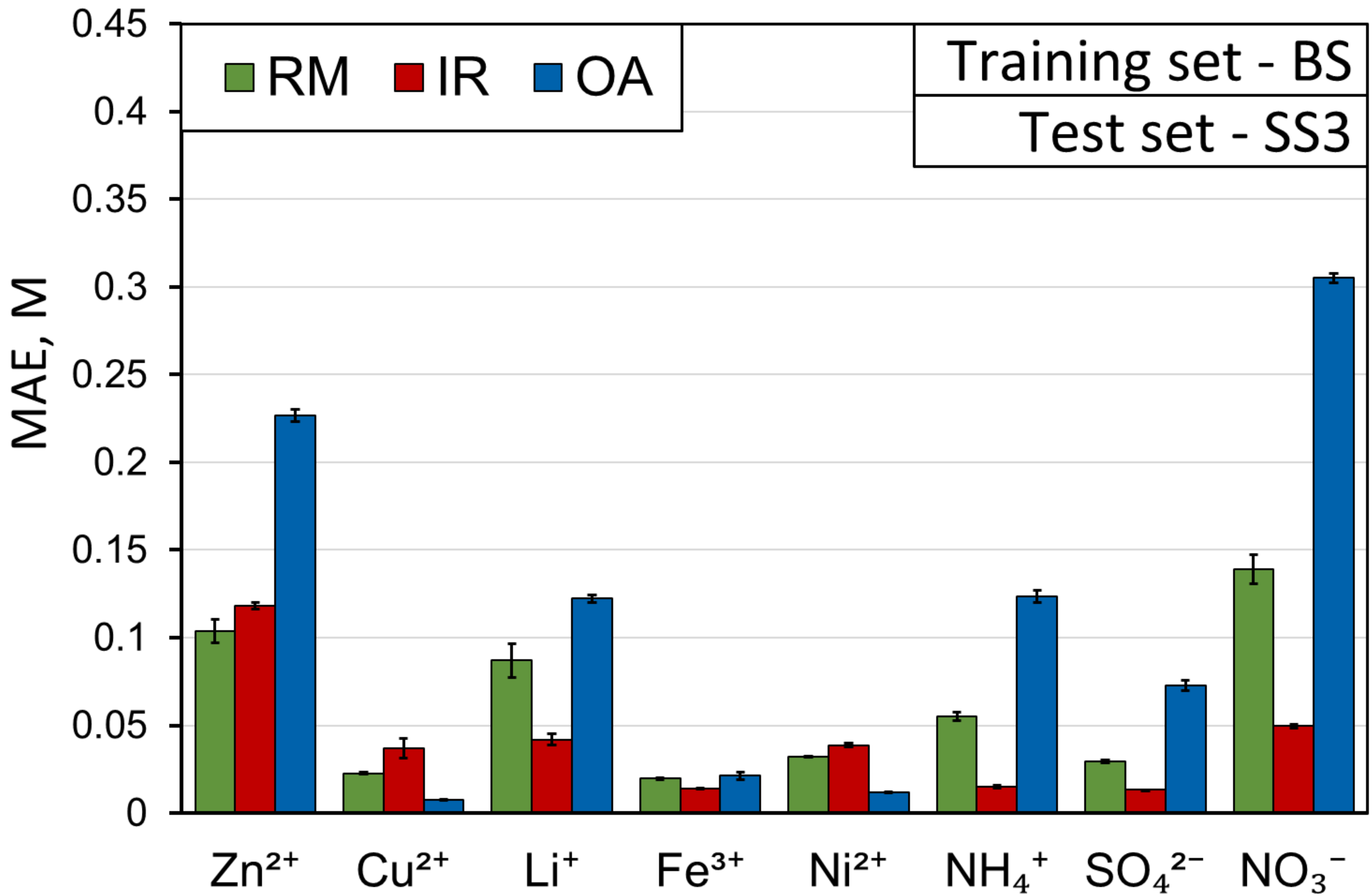

RM
IR
OA
Training set - BS
Test set - SS1
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn^{2+}
Cu^{2+}
Li^{+}
Fe^{3+}
Ni^{2+}
NH_4^{+}
SO_4^{2-}
NO_3^{-}

Results (1)



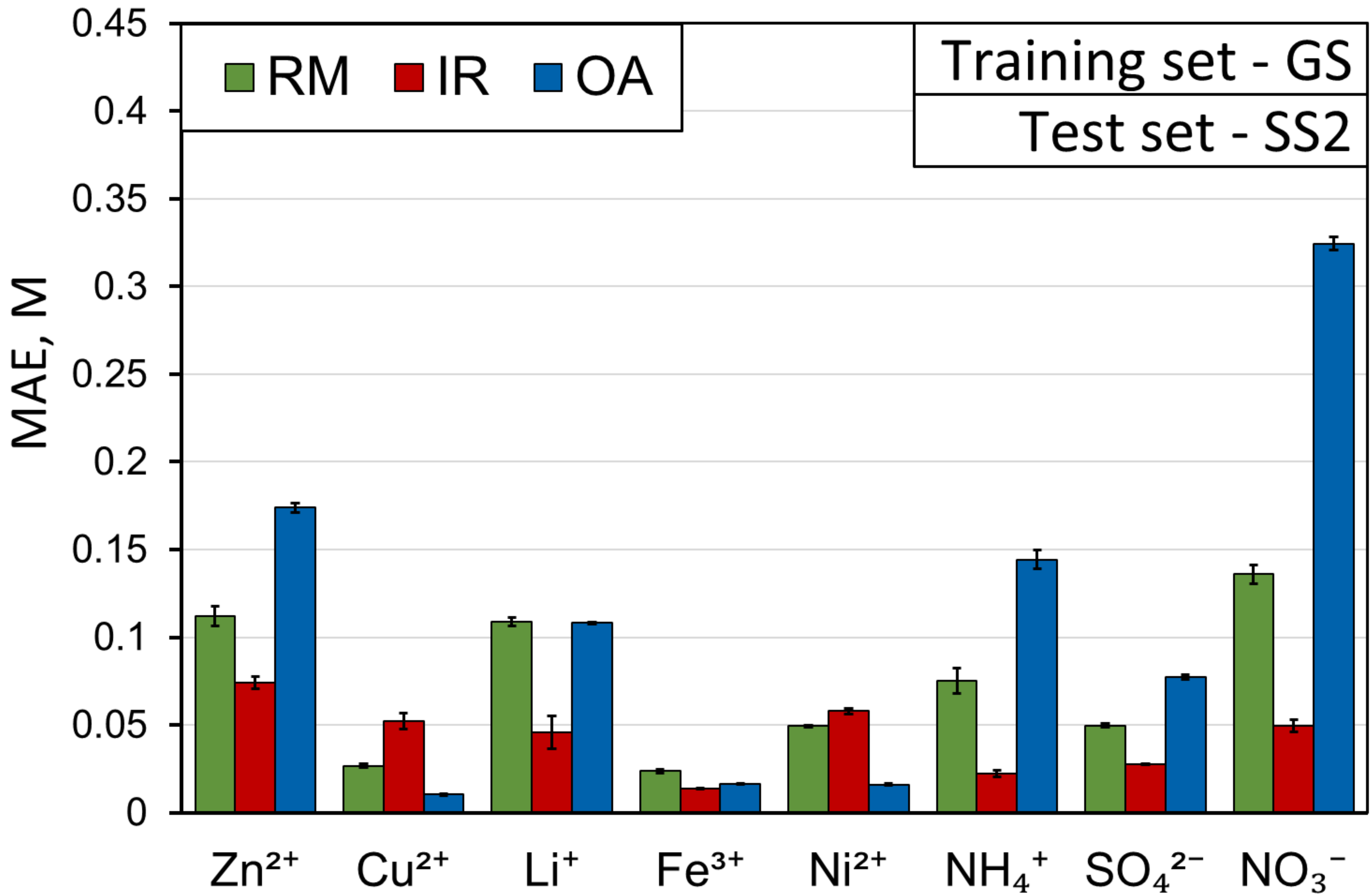
RM
IR
OA
Training set - BS
Test set - SS2
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn^{2+}
Cu^{2+}
Li^{+}
Fe^{3+}
Ni^{2+}
NH_4^{+}
SO_4^{2-}
NO_3^{-}

Results (1)



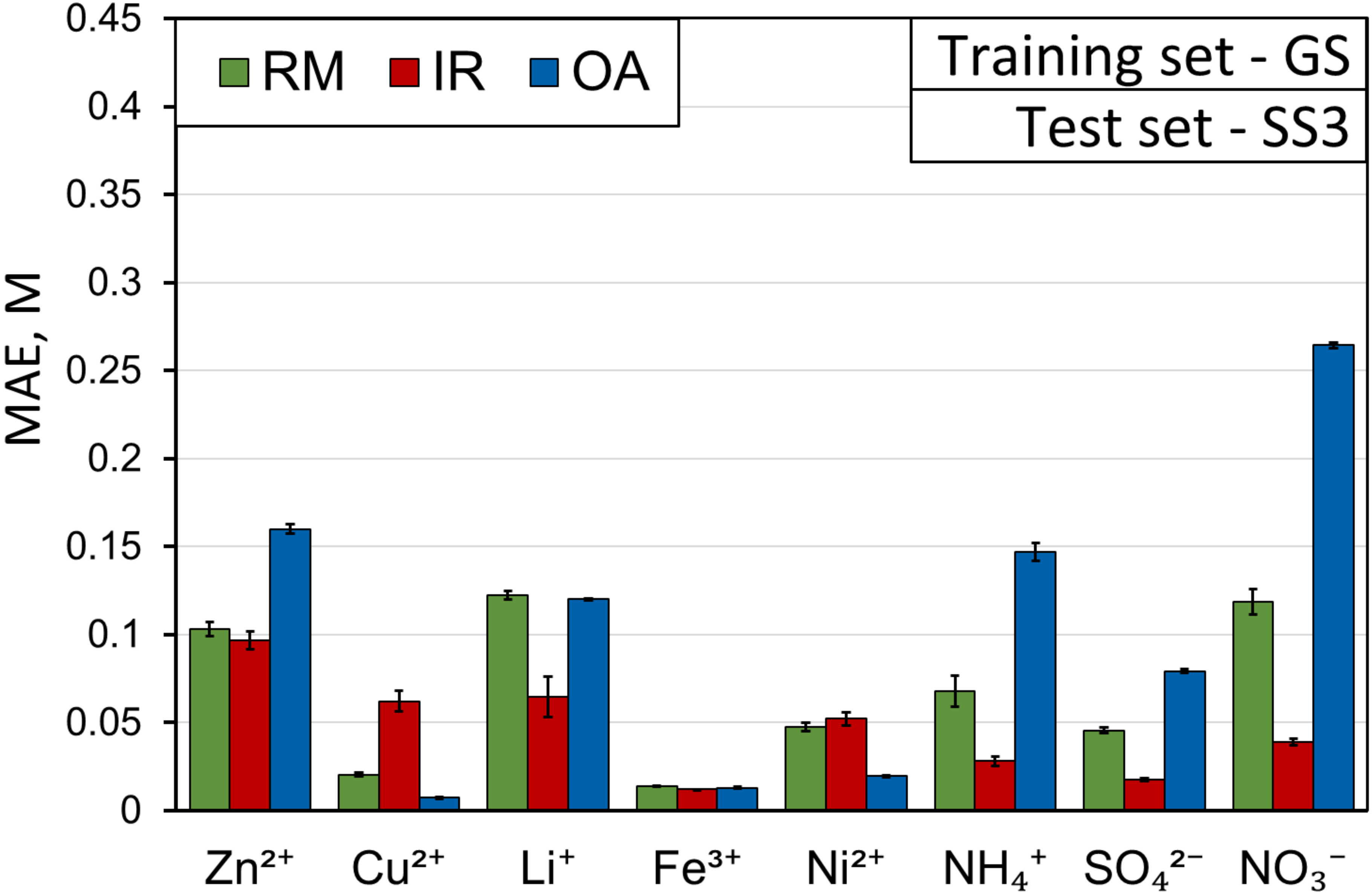
RM
IR
OA
Training set - BS
Test set - SS3
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn^{2+}
Cu^{2+}
Li^{+}
Fe^{3+}
Ni^{2+}
NH_4^{+}
SO_4^{2-}
NO_3^{-}

Results (2)



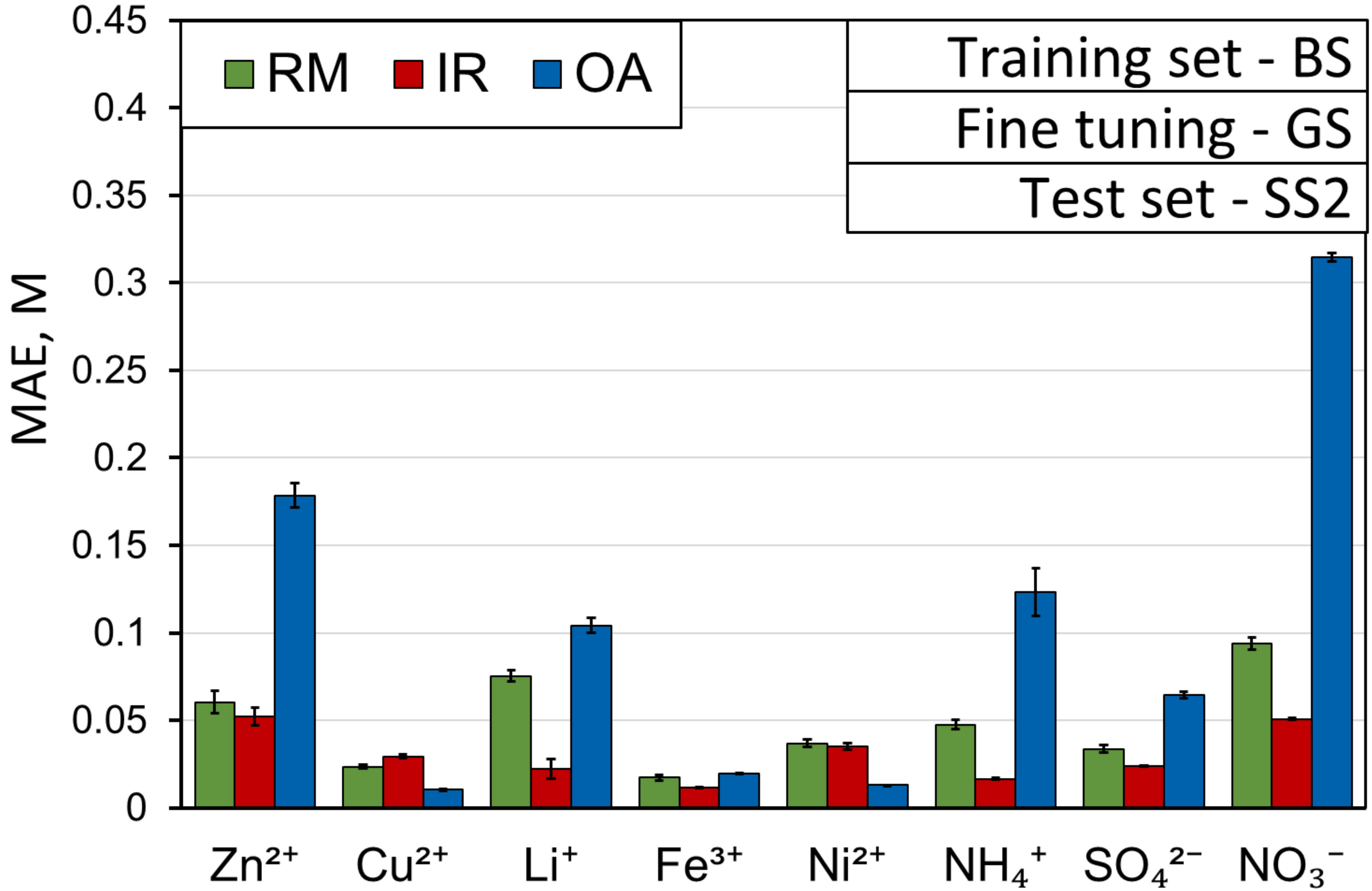
RM
IR
OA
Training set - GS
Test set - SS2
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn^{2+}
Cu^{2+}
Li^{+}
Fe^{3+}
Ni^{2+}
NH_4^{+}
SO_4^{2-}
NO_3^{-}

Results (2)



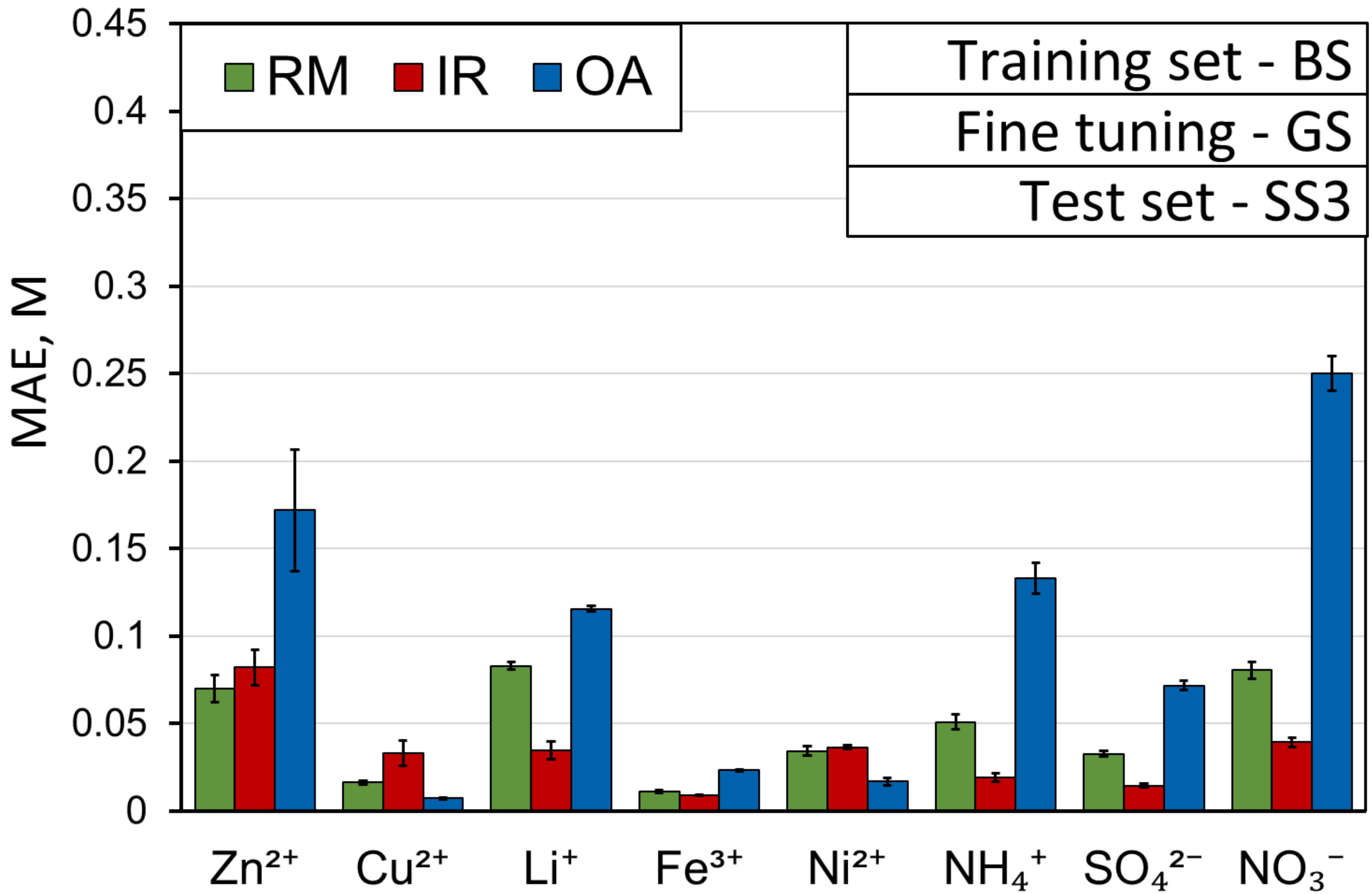
RM
IR
OA
Training set - GS
Test set - SS3
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn²⁺
Cu²⁺
Li⁺
Fe³⁺
Ni²⁺
NH₄⁺
SO₄²⁻
NO₃⁻

Results (3)



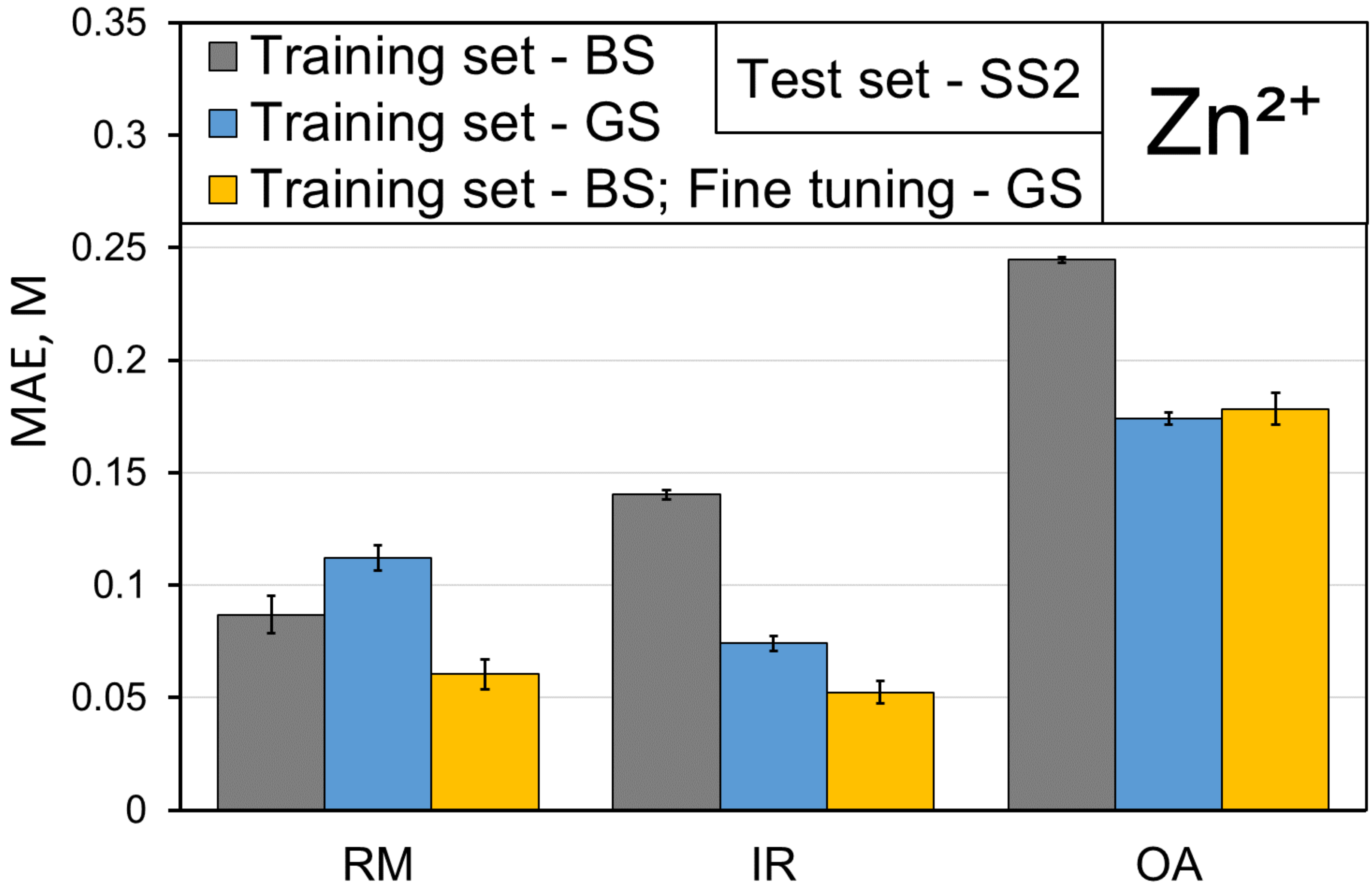
RM
IR
OA
Training set - BS
Fine tuning - GS
Test set - SS2
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn²⁺
Cu²⁺
Li⁺
Fe³⁺
Ni²⁺
NH₄⁺
SO₄²⁻
NO₃⁻

Results (3)



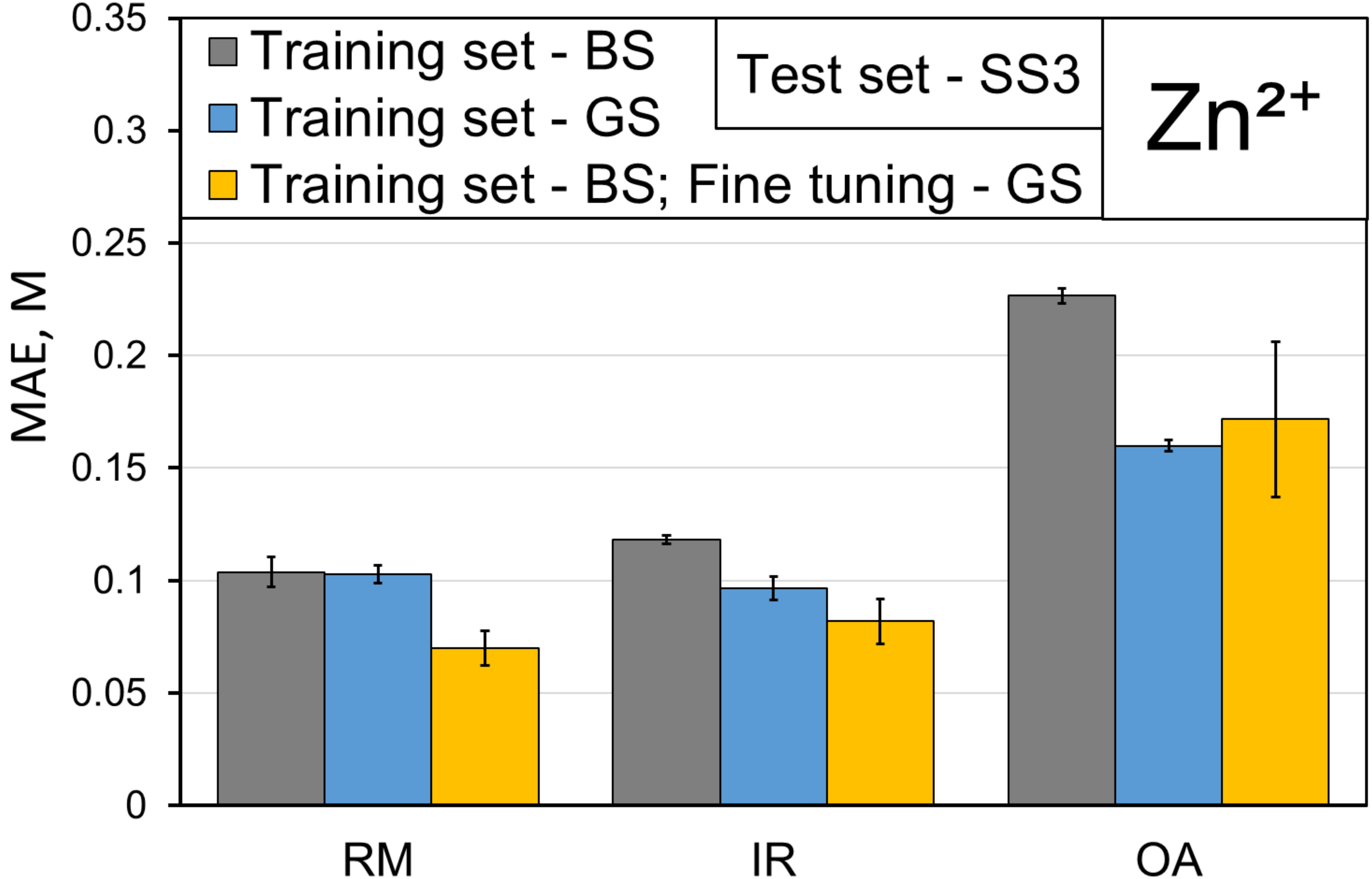
RM
IR
OA
Training set - BS
Fine tuning - GS
Test set - SS3
MAE, M
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Zn²⁺
Cu²⁺
Li⁺
Fe³⁺
Ni²⁺
NH₄⁺
SO₄²⁻
NO₃⁻

Results (4)



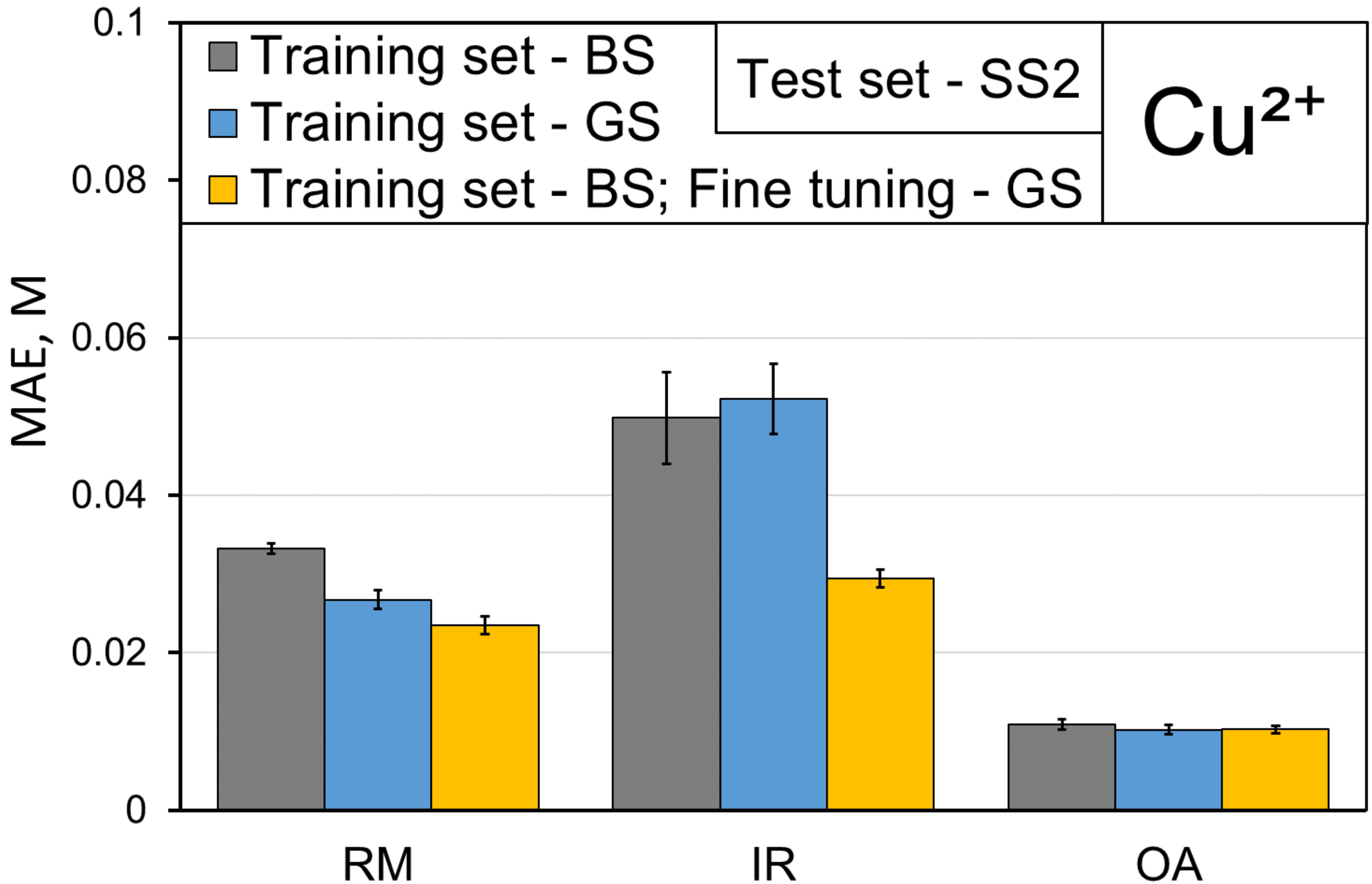
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
MAE, M
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
Zn2+
RM
IR
OA

Results (4)



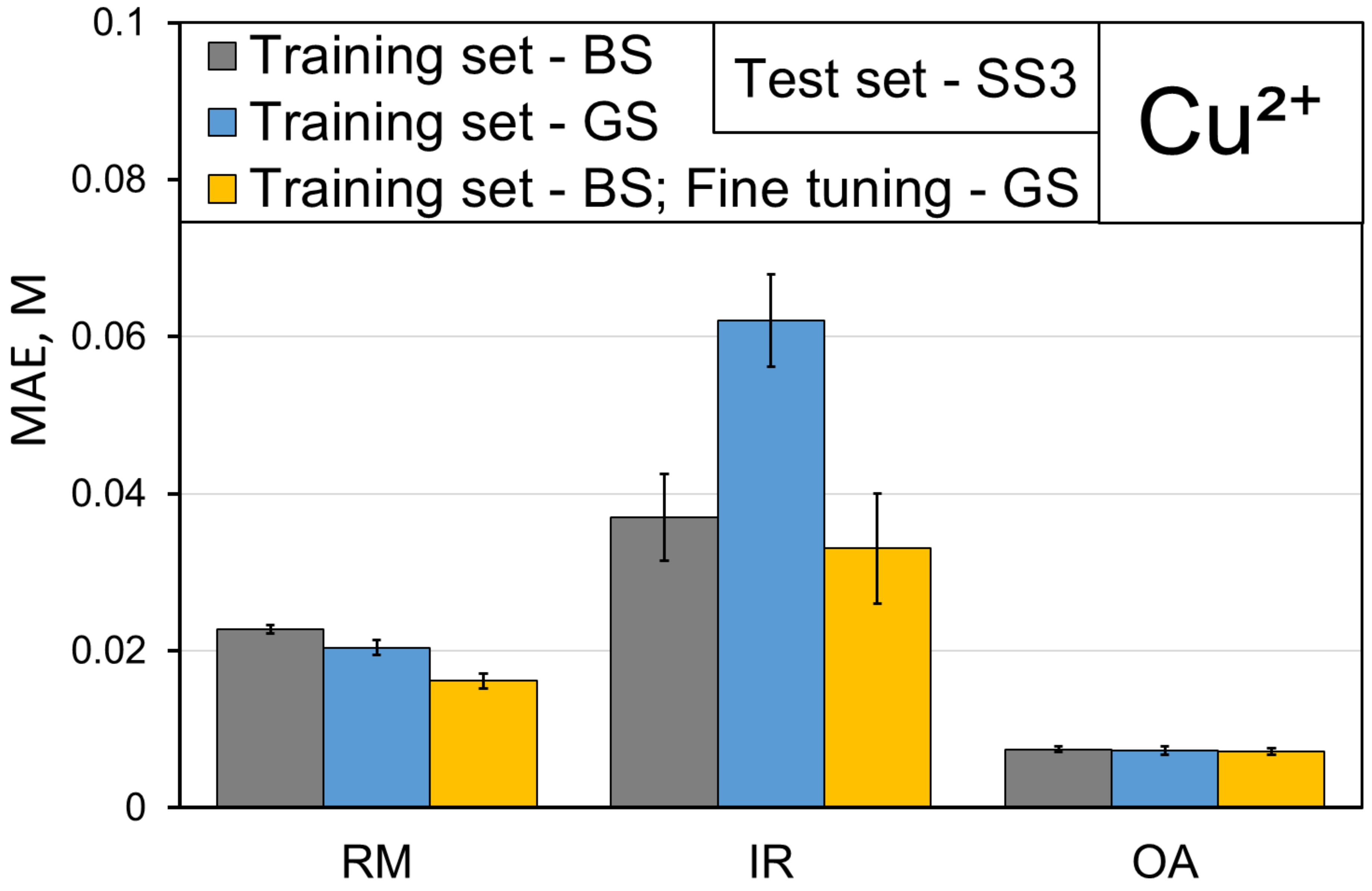
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
Zn²⁺
MAE, M
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
RM
IR
OA

Results (4)



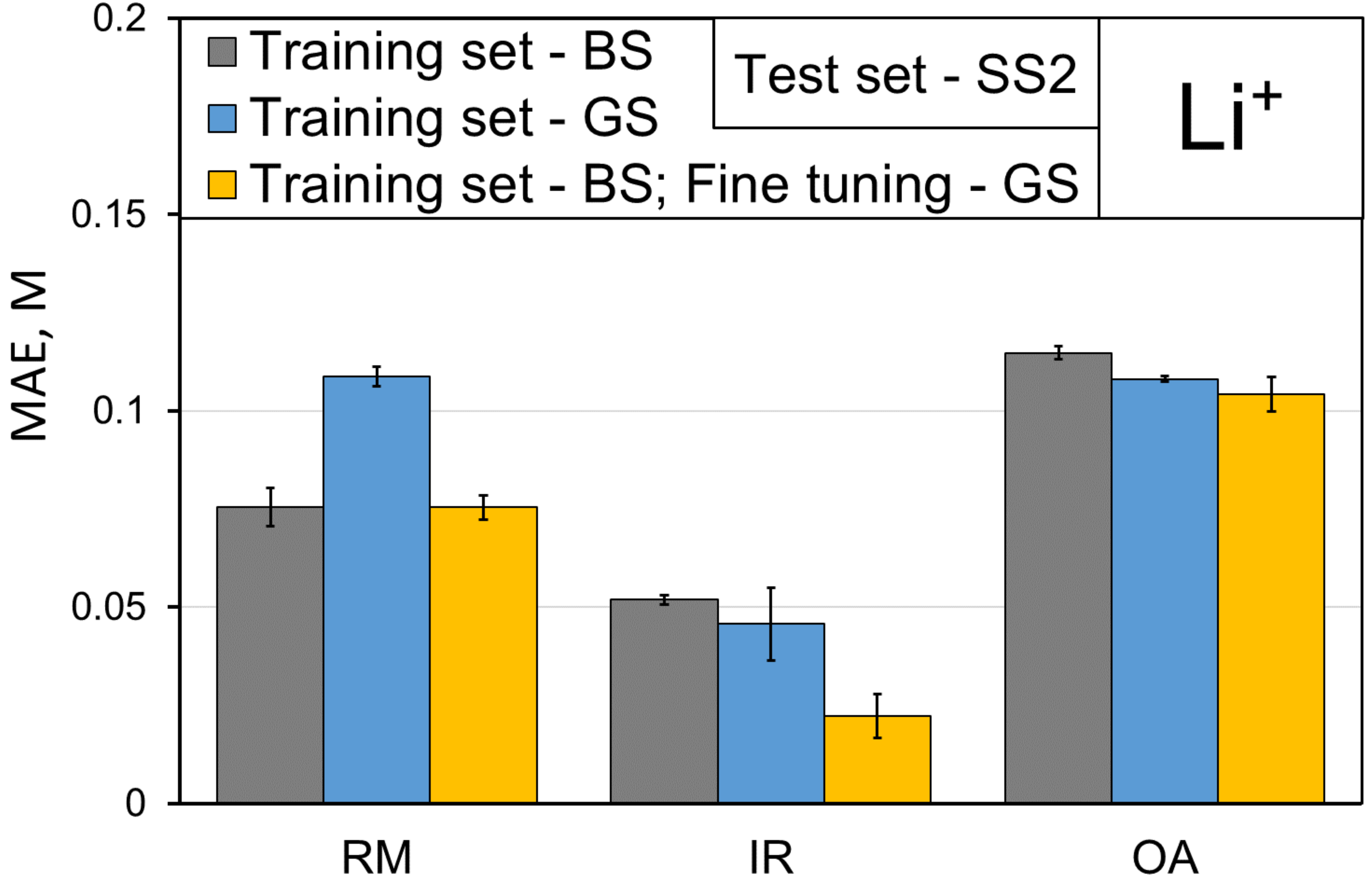
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
Cu2+
MAE, M
0.1
0.08
0.06
0.04
0.02
0
RM
IR
OA

Results (4)



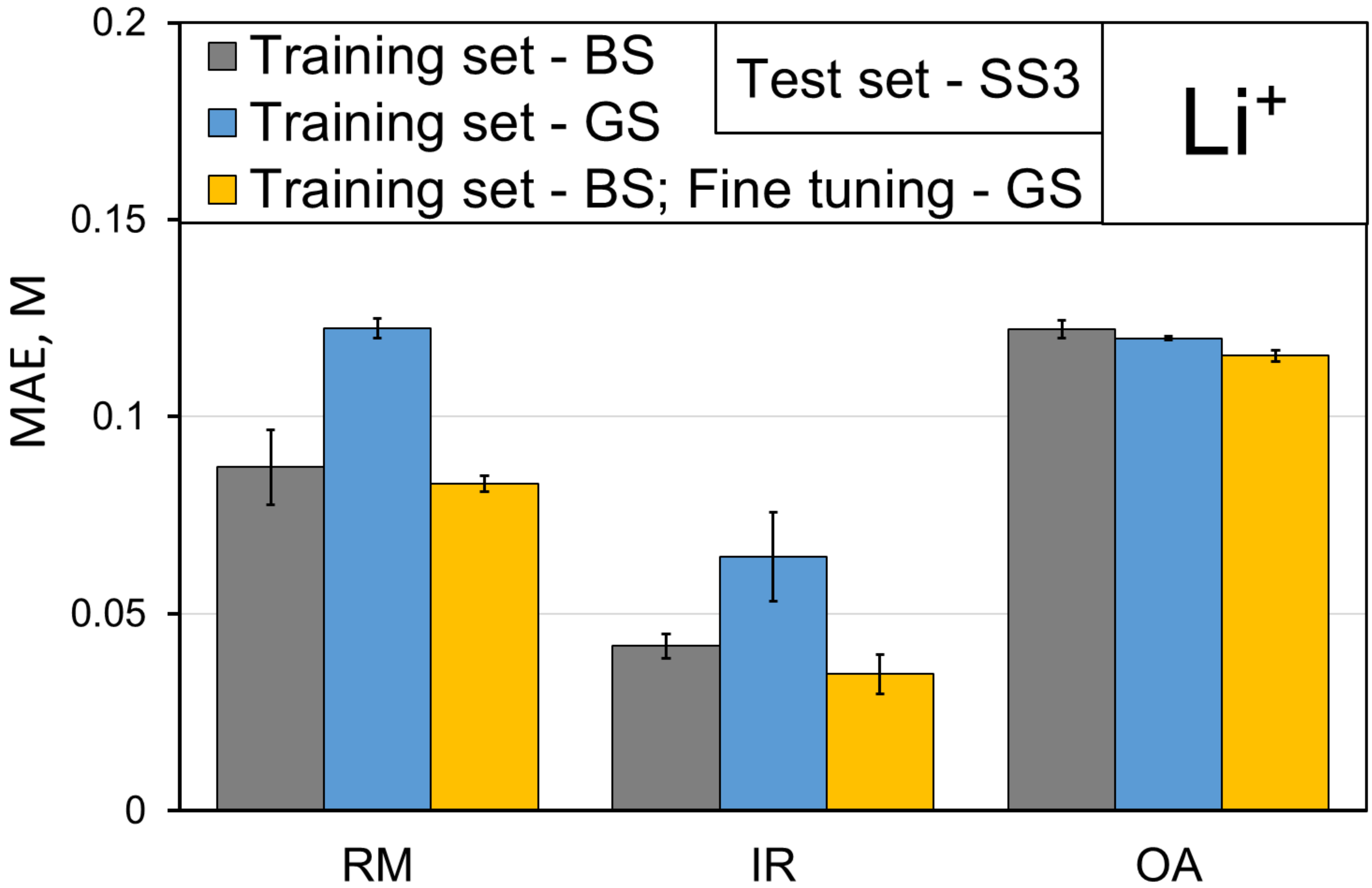
0.1
0.08
0.06
0.04
0.02
0
MAE, M
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
Cu2+
RM
IR
OA

Results (4)



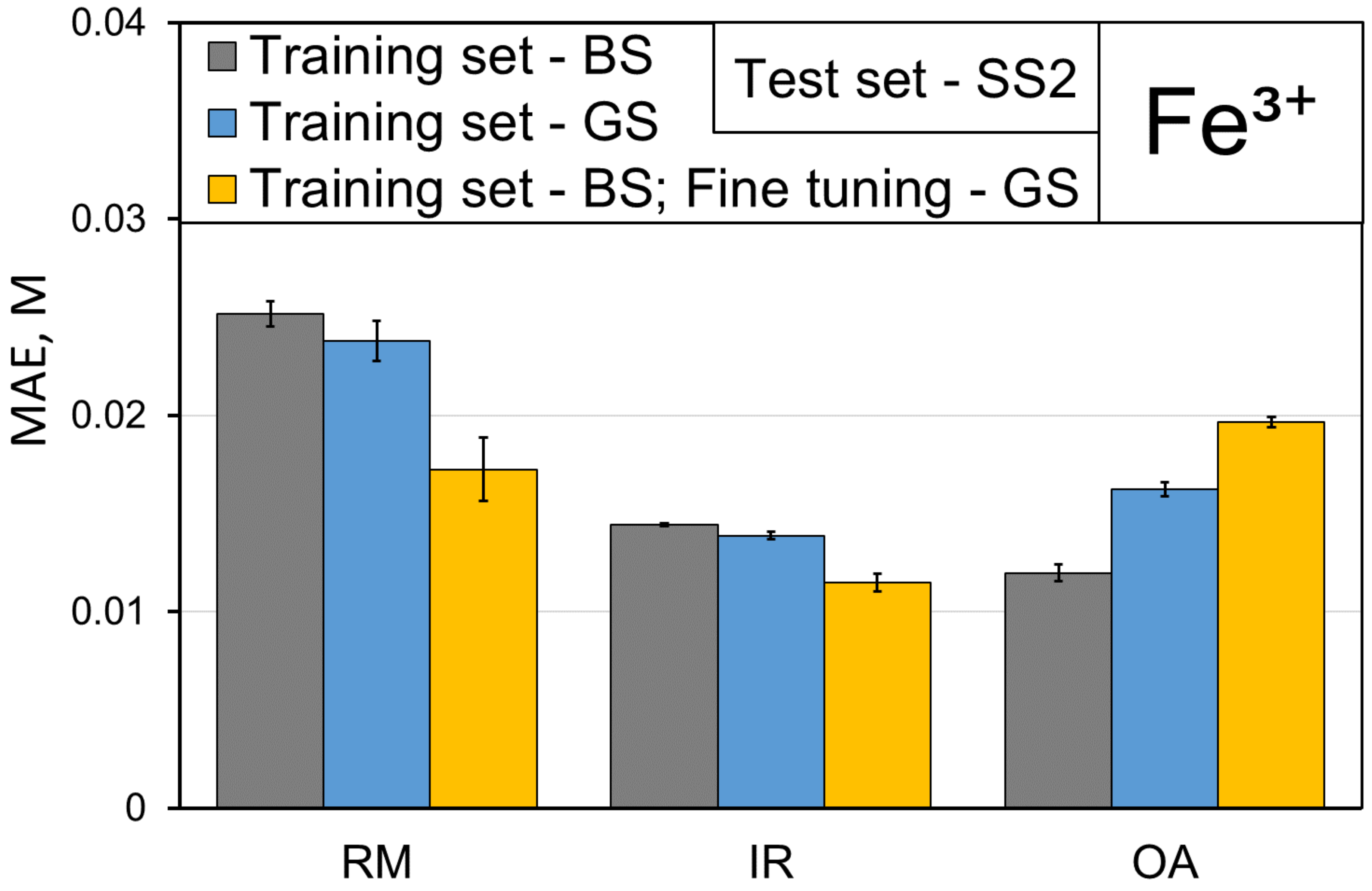
0.2
0.15
0.1
0.05
0
MAE, M
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
Li+
RM
IR
OA

Results (4)



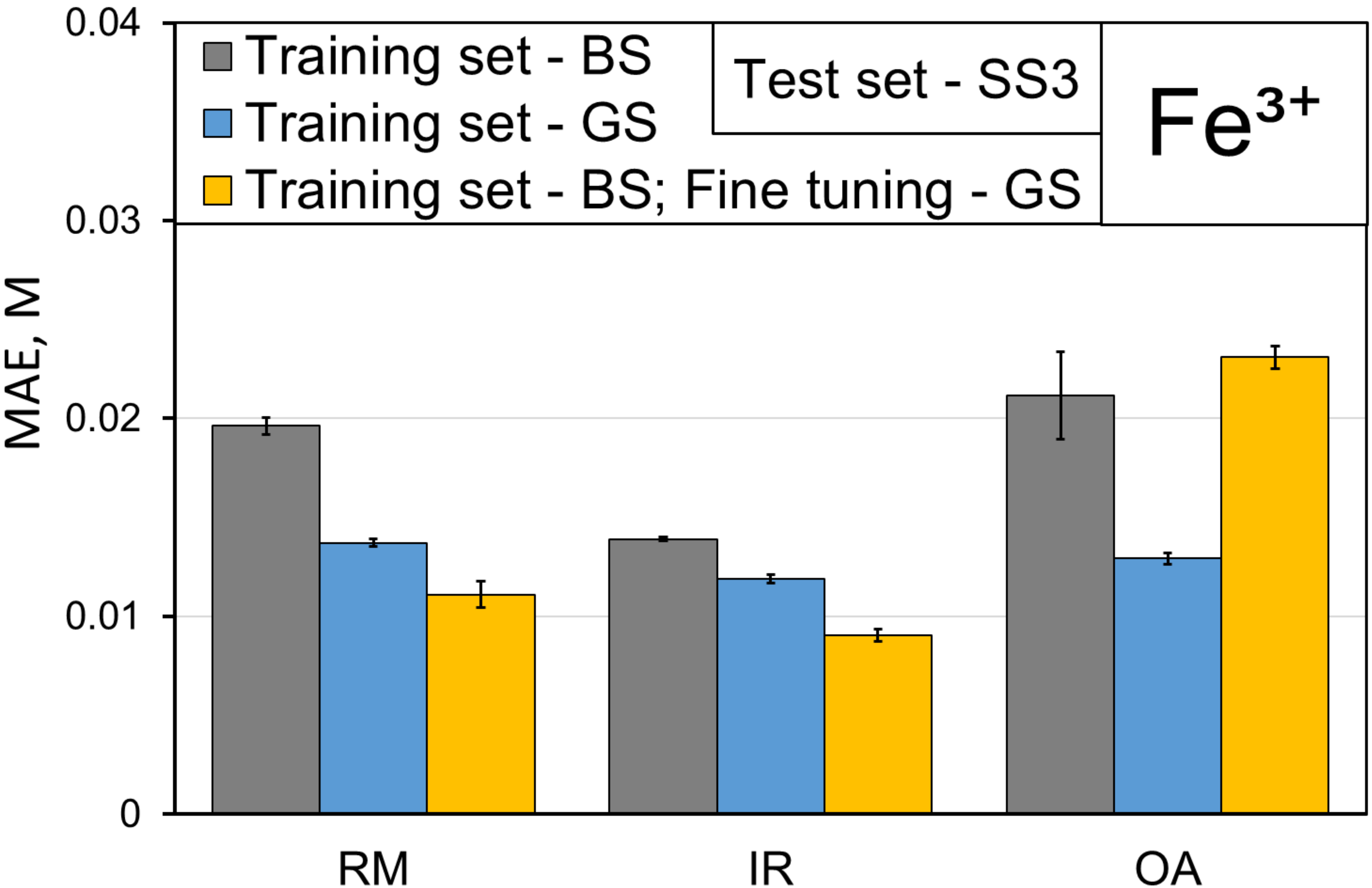
0.2
0.15
0.1
0.05
0
MAE, M
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
Li+
RM
IR
OA

Results (4)



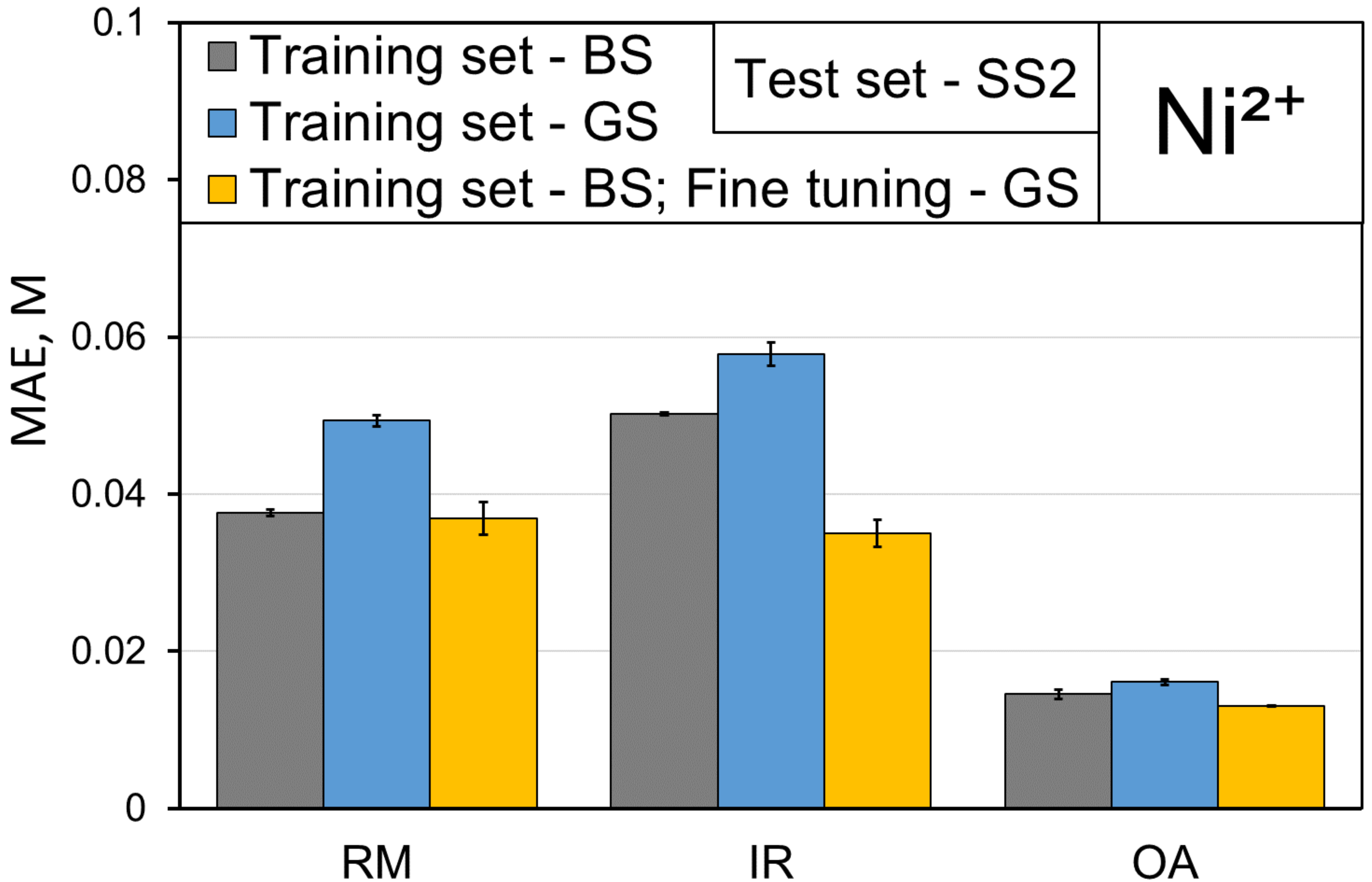
0.04
0.03
0.02
0.01
0
MAE, M
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
Fe^{3+}
RM
IR
OA

Results (4)



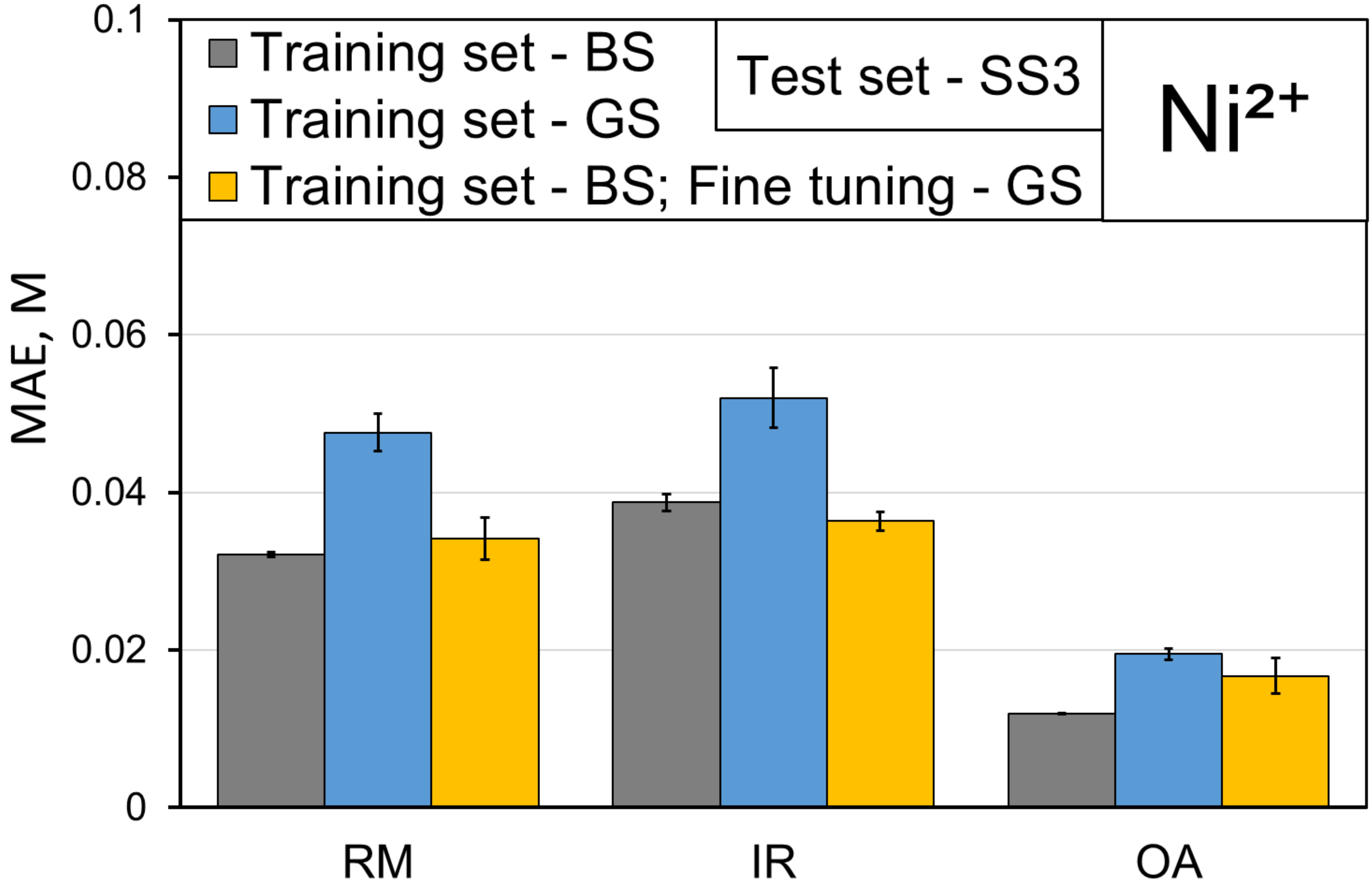
0.04
0.03
0.02
0.01
0
MAE, M
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
Fe3+
RM
IR
OA

Results (4)



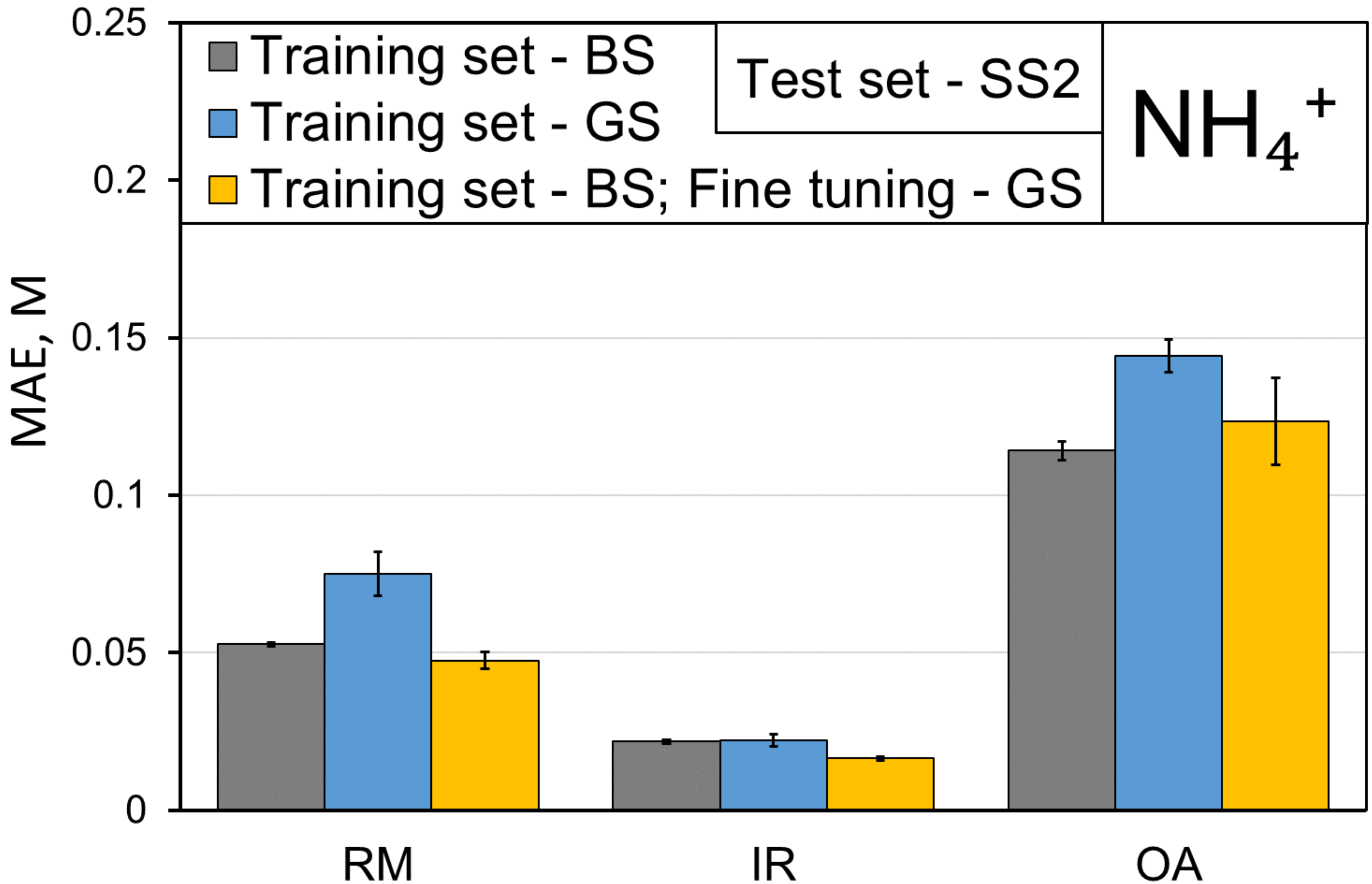
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
Ni2+
MAE, M
0.1
0.08
0.06
0.04
0.02
0
RM
IR
OA

Results (4)



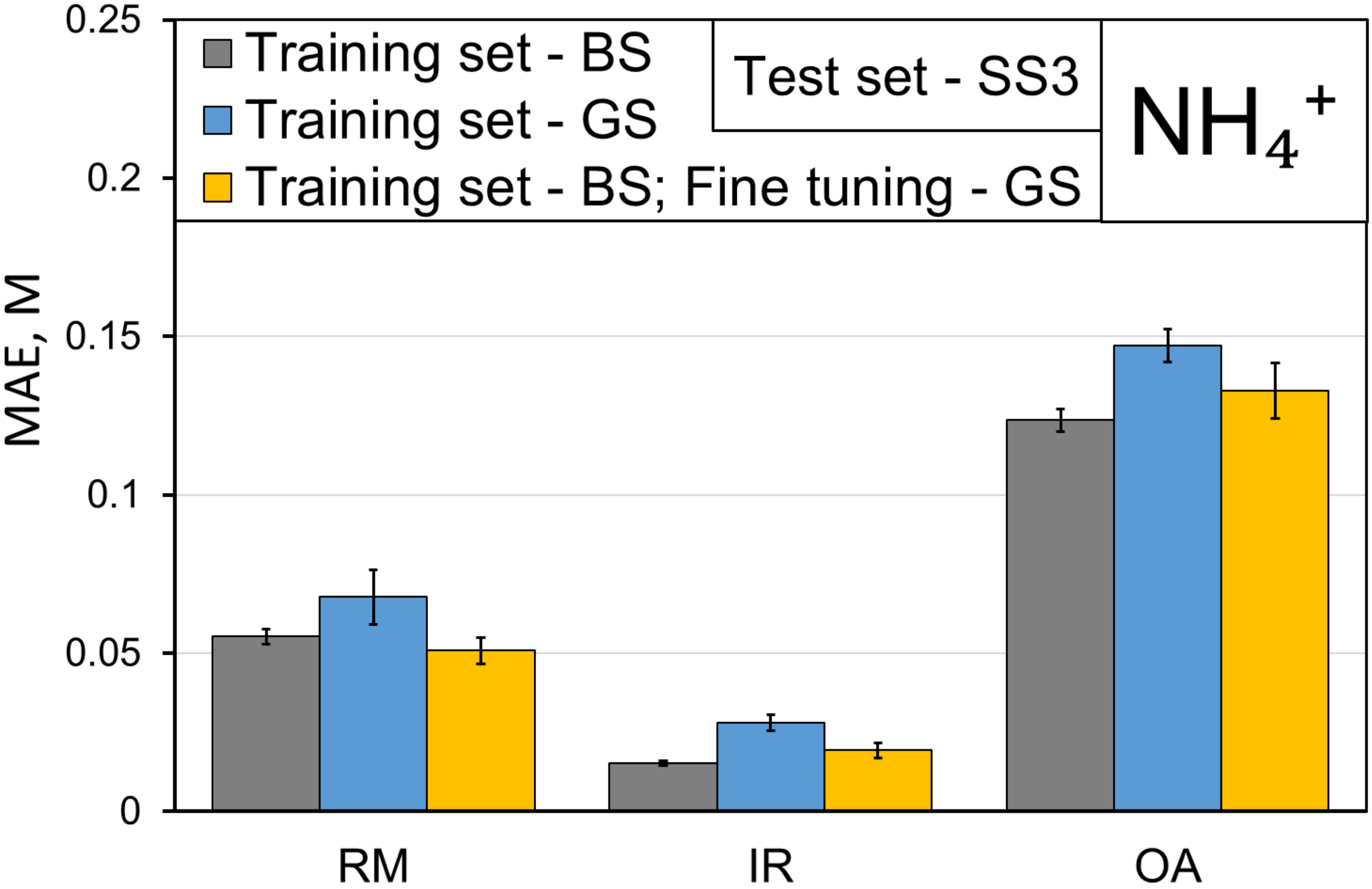
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
Ni2+
MAE, M
0.1
0.08
0.06
0.04
0.02
0
RM
IR
OA

Results (4)



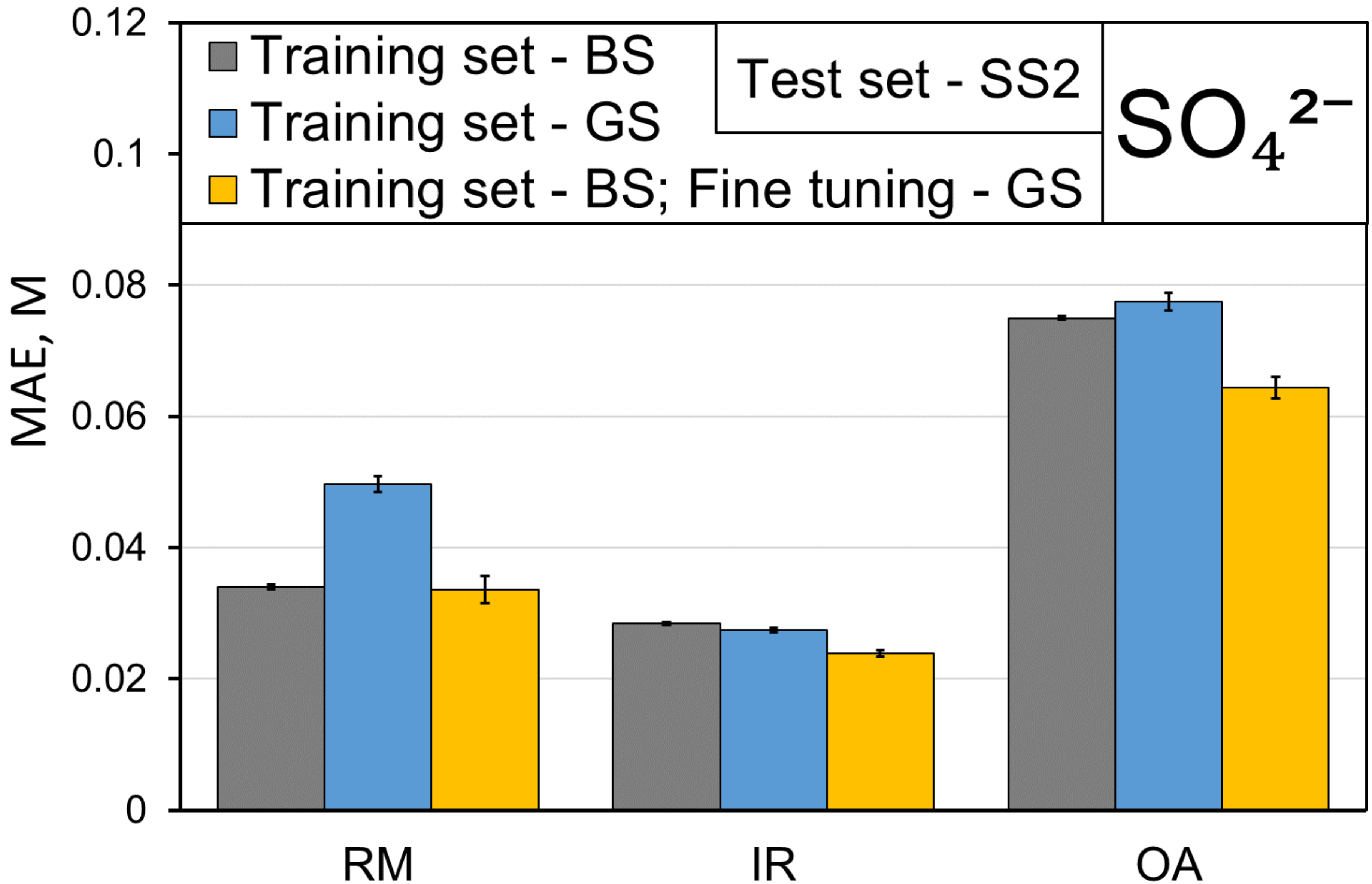
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
NH_4^+
MAE, M
0.25
0.2
0.15
0.1
0.05
0
RM
IR
OA

Results (4)



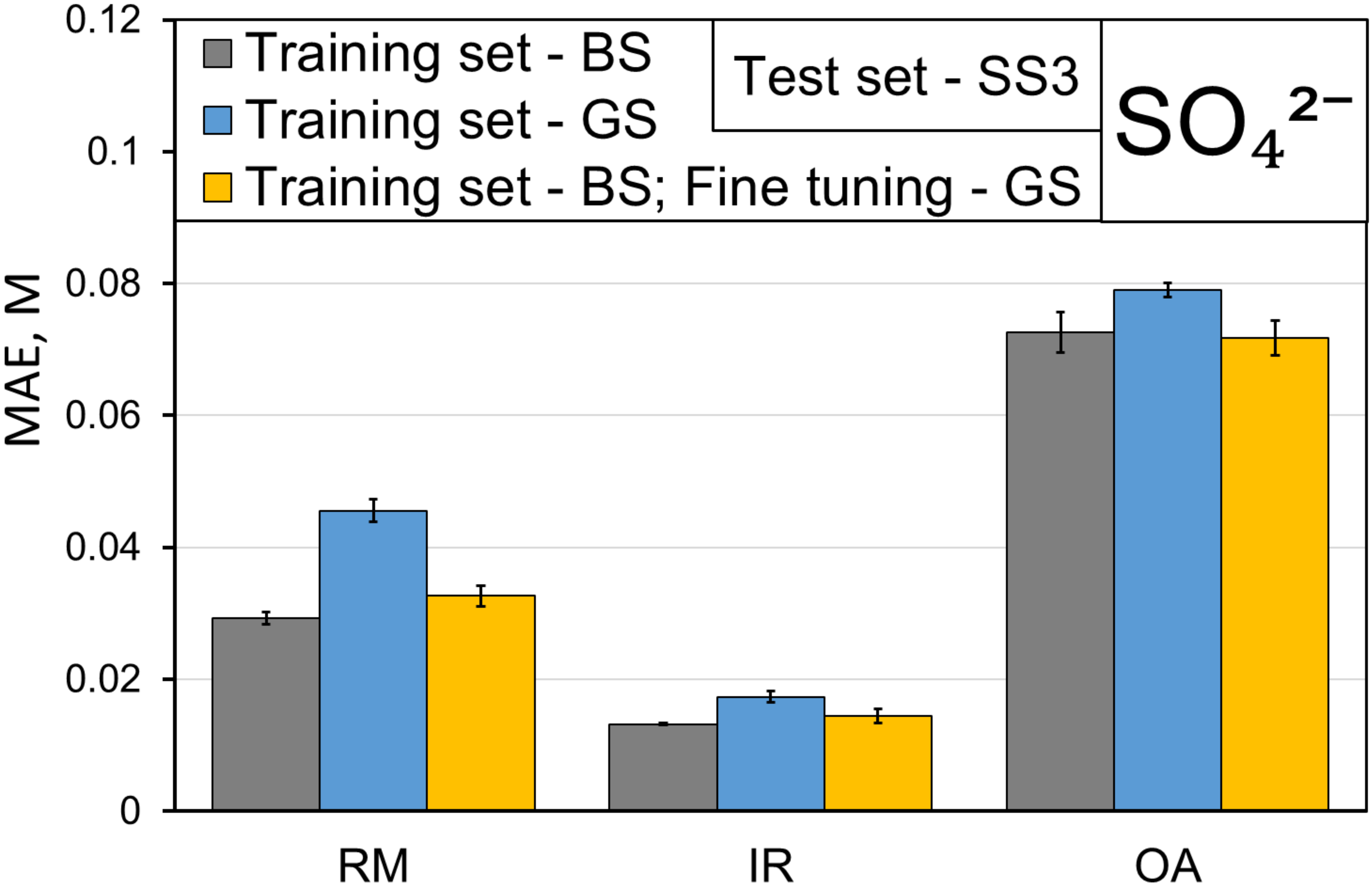
Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
NH4+
MAE, M
0.25
0.2
0.15
0.1
0.05
0
RM
IR
OA

Results (4)



Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
SO_4^{2-}
MAE, M
0.12
0.1
0.08
0.06
0.04
0.02
0
RM
IR
OA

Results (4)

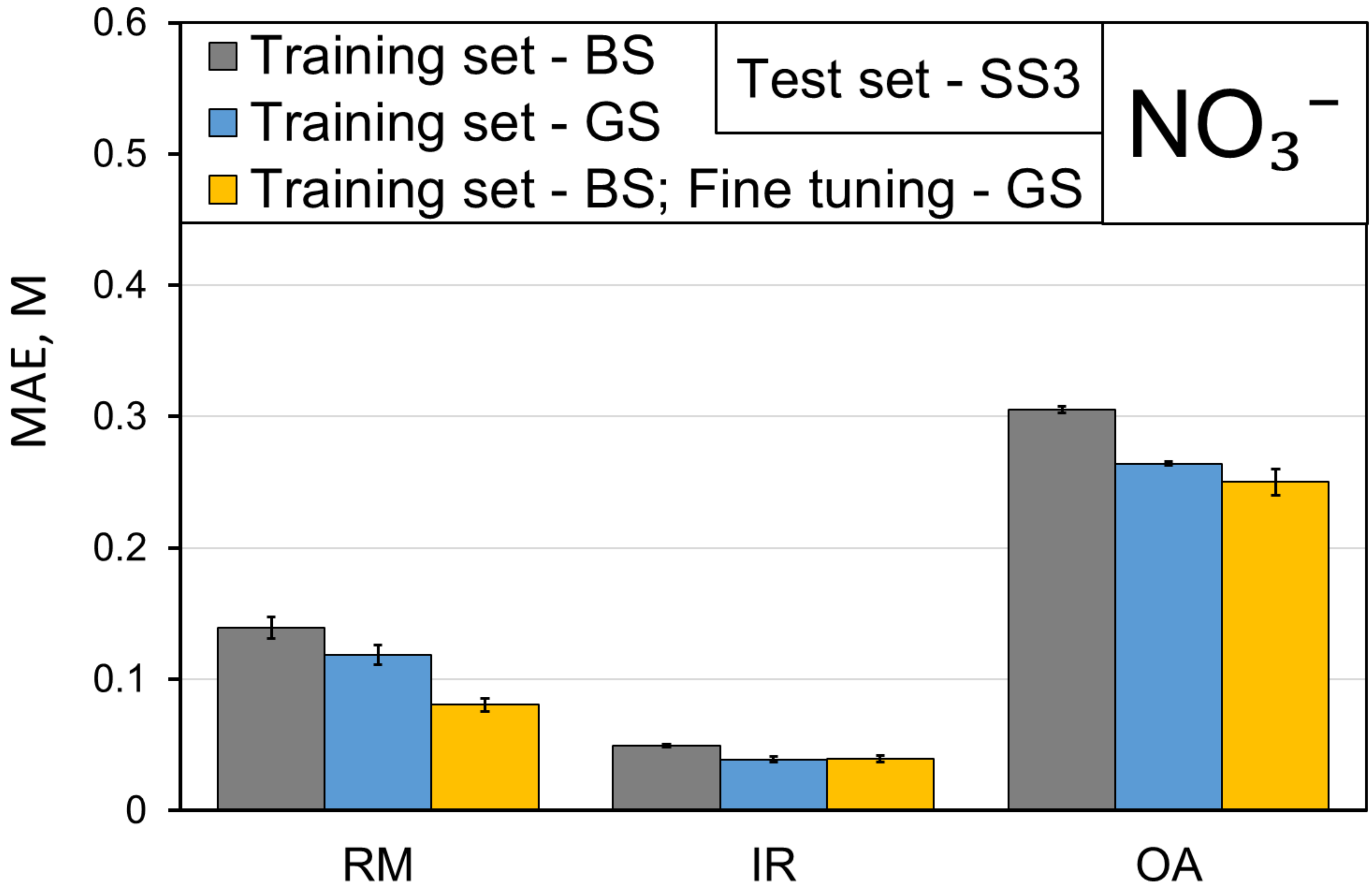


Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
SO_4^{2-}
MAE, M
0.12
0.1
0.08
0.06
0.04
0.02
0
RM
IR
OA

Results (4)

Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS2
NO_3^-
MAE, M
0.6
0.5
0.4
0.3
0.2
0.1
0
RM
IR
OA

Results (4)



Training set - BS
Training set - GS
Training set - BS; Fine tuning - GS
Test set - SS3
NO_3^-
MAE, M
0.6
0.5
0.4
0.3
0.2
0.1
0
RM
IR
OA

Conclusions

1) NN trained on distilled water data demonstrate high performance on the same type of data

2) Its performance degrades several fold when applied to river water data

3) NN trained on river water data only gives error of the same order, but it is much less stable

4) Use of transfer learning (taking NN pre-trained on distilled water data, fine tuning it on river water data) shows stable better results

5) Integration of data of several spectroscopy types should be tested together with transfer learning (future study)

Thank you for your attention